

# Philips Technical Review

DEALING WITH TECHNICAL PROBLEMS  
RELATING TO THE PRODUCTS, PROCESSES AND INVESTIGATIONS OF  
THE PHILIPS INDUSTRIES

## THE PRODUCTION AND MEASUREMENT OF ULTRA-HIGH VACUA

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*In order to produce ultra high vacua (i.e. pressures lower than  $10^{-7}$  mm Hg) it was the practice until recently to work with a closed system and to use in the last phase of the pumping process some form of "internal pump", e.g. a getter, an ionization gauge or both. The article below shows that it is also possible to attain vacua considerably better than  $10^{-7}$  mm Hg with a normal mercury diffusion pump, which has the advantage of a considerably greater pumping speed. The mercury diffusion pump must, for this purpose, form part of a specially designed vacuum system.*

Recent years have seen a marked growth of interest in ultra-high vacuum technique. The reason is that the extremely low pressures offer interesting possibilities for the investigation of surface properties, e.g. for measuring contact potentials and studying adsorption phenomena. Only when the pressure of the ambient gas atmosphere is sufficiently low can a completely clean surface remain clean for a time long enough to allow experiments to be carried out on it. At a pressure of  $10^{-7}$  mm Hg a clean surface may be completely covered by a monolayer in a matter of 10 seconds; even at this low pressure the number of molecules from the gas atmosphere incident per second on one  $\text{cm}^2$  at room temperature is still  $4 \times 10^{13}$ . A reduction of the pressure by, say, a factor of 1000 increases the time available for experiments by the same factor.

A few years ago it was thought that pressures in the range of  $10^{-7}$  to  $10^{-10}$  mm Hg could hardly be reached. A pressure of  $10^{-7}$  mm Hg was regarded as particularly low, and the fact that it was not possible to reach lower pressures was attributed to incomplete mastery of pumping technique. In 1947, however, Nottingham<sup>1)</sup> pointed out that the measuring instrument used in this pressure range, the ionization gauge, might not be capable of giving a true reading of such extremely low pressures. This

investigator suggested that every ionization gauge has a certain minimum indication which depends on the design of the gauge; with the instruments used at that time this indication corresponded to a pressure of  $10^{-7}$  to  $10^{-8}$  mm Hg. In 1950 and 1951, manometer designs were proposed by Bayard and Alpert<sup>2)</sup>, by Lander<sup>3)</sup> and by Metson<sup>4)</sup>, which offered an appreciable extension of the measuring range. In 1953 and 1954 Alpert<sup>5)</sup> and Alpert and Buritz<sup>6)</sup> described in two detailed articles a technique by which it was possible to produce and reliably measure vacua in the range from  $10^{-9}$  to  $10^{-10}$  mm Hg.

In the present article we shall describe a pumping system by means of which pressures in the range from  $10^{-10}$  to  $10^{-12}$  mm Hg can be achieved, as well as an ionization gauge for measuring these pressures. First, however, we shall deal with the problem of lowering the minimum reading of an ionization gauge (and hence the smallest measurable pressure) after which we shall discuss Alpert's method of producing a pressure of  $10^{-10}$  mm Hg, and mention some general aspects of the pumping process at extremely low pressures. From these aspects we shall derive the principles on which the design of our pumping system is based.

<sup>1)</sup> W. B. Nottingham, information communicated to D. Alpert (see <sup>5)</sup>).

<sup>2)</sup> R. T. Bayard and D. Alpert, Rev. sci. Instr. **21**, 571, 1950.

<sup>3)</sup> J. J. Lander, Rev. sci. Instr. **21**, 672, 1950.

<sup>4)</sup> G. H. Metson, Brit. J. appl. Phys. **2**, 46, 1951.

<sup>5)</sup> D. Alpert, J. appl. Phys. **24**, 860, 1953.

<sup>6)</sup> D. Alpert and R. S. Buritz, J. appl. Phys. **25**, 202, 1954.



### *Ionization gauges for ultra-high vacua*

In the pressure range with which we are concerned the only possible measuring instrument is the ionization gauge. The normal design consists of a cathode (usually a tungsten filament) situated in the middle of the electrode system, surrounded coaxially by the anode, in the form of a cylindrical wire helix, and the ion collector, also in the form of a helix; see *fig. 1*. In some cases an additional electrode is

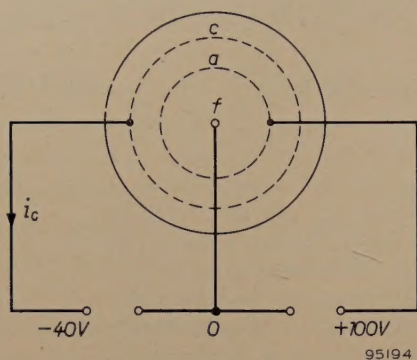


Fig. 1. Principle of the ionization gauge. The electrons emitted by cathode *f* are accelerated in the field between *f* and the anode *a* (the latter in the form of a cylindrical wire helix) giving them a kinetic energy high enough to enable them to ionize gas molecules. The electrons finally arrive at *a*. The positive ions formed in the space between *a* and the collector *c* (of the same form as the anode) move to the collector. Within a certain pressure range, the current  $i_c$  in the collector circuit is proportional to the pressure.

introduced between cathode and anode for the purpose of stabilizing the electron current<sup>7)</sup>. The instrument functions as follows. The electrons emitted by the cathode are accelerated in the field between cathode and anode until their kinetic energy is high enough for them to be capable of ionizing gas molecules. The ions formed in the space between cathode and anode play no part in the measurement; the positive ions, however, which are formed in the space between anode and collector, are attracted towards the collector, which is at a lower potential than the anode. The resultant current flowing in the collector resistance is proportional, over a wide range of pressures, to the number of molecules present per unit volume, and is hence proportional to the pressure. This proportionality is lost, however, when pressures in the region of  $10^{-8}$  mm Hg are reached. In fact, the current appears to be unable to drop below a specific value no matter how low the pressure may be.

What is the cause of this residual reading? Nottingham<sup>1)</sup> assumed that the residual current

is not due to positive ions impinging on the collector, but to photoelectrons liberated from the collector as a result of soft X-rays which are generated when electrons from the cathode strike the anode. In order to be able to measure pressures lower than about  $10^{-8}$  mm Hg it is therefore necessary to reduce this "X-ray effect". Gauges in which this has been achieved are reported in the publications<sup>2), 3)</sup> and<sup>4)</sup> mentioned above. The instrument built by Bayard and Alpert<sup>2)</sup> is the simplest in design and therefore most commonly used. In this type of ionization gauge the ion collector is a thin wire mounted centrally in the cylindrical anode. The cathode is outside the anode. The sensitivity with this electrode arrangement is about the same as with the old design but there is now substantially less electron emission from the ion collector. In this way the "X-ray limit" is shifted to a pressure which is 100-1000 times smaller than in the old version. The ionization gauge which we have built, and which will be described in this article, is also of the type with central collector.

### *Alpert's method of producing extremely low pressures*

The technique described by Alpert<sup>5)</sup> for obtaining reliable vacua in the range of  $10^{-10}$  mm Hg is based on a property common to every ionization gauge and which, depending on the circumstances, may be either an advantage or a disadvantage. This is the property of the gauge to adsorb gas more or less permanently on the glass envelope and on certain electrodes. In most cases the glass envelope of the gauge acquires a charge such that positive ions are attracted to the glass where they are neutralized and sorbed as molecules. The same can happen on the electrodes when the latter have the appropriate potential. At an electron current smaller than about 0.01 mA this pumping action is usually slight, but it is quite pronounced if the electron current is 0.1 mA or more. Its effect on pressure measurements can be distorting in view of the fact that the ion gauge must as a rule be connected to the vessel via a tube. Between the ends of this tube a noticeable difference in pressure can arise, and the gauge reading is then no longer a direct measure of the pressure in the vessel. The pumping action of the ion gauge can, however, also be turned to advantage for the purpose of cleaning up the small amounts of gas which, even after careful outgassing of walls and components, may still be liberated inside the system. By this means the pressure can be reduced to a low value and maintained at that value.

<sup>7)</sup> An instrument of this type was described earlier in this Review by E. Bouwmeester and N. Warmoltz: Philips tech. Rev. 17, 121, 1955/56; see also Appl. sci. Res. B 2, 273, 1951.



The pump system developed by Alpert on this principle consists of an oil-diffusion pump, capable of reducing the pressure in the system to be evacuated to a value of about  $10^{-7}$  mm Hg, an all-metal valve which isolates the pump from the system, and the ionization gauge. If, after the requisite pressure has been reached, it is required to introduce a certain gas into the system, use can be made of a second metal valve, which connects the system to a reservoir. With the first valve open and the pressure in the system having been reduced to about  $10^{-7}$  mm Hg by the diffusion pump, the entire system is placed in a "bake-out" furnace (with the pump attached, but outside the furnace) and heated for six to eight hours at a temperature of about  $450^\circ\text{C}$ . This having been done, the gauge electrodes are outgassed by electron bombardment. The valve is then closed and the ionization gauge set in operation with an electron current of 10 mA. The pressure is now observed to fall, and within an hour it reaches a value in the range from  $10^{-9}$  to  $10^{-10}$  mm Hg.

#### *Factors governing the lowest attainable pressure*

We shall now turn to the principles underlying our pump system, which we shall derive from a general consideration of the pumping process in the range of low and extremely low pressures and from a closer analysis of Alpert's method.

When a vessel is evacuated one generally notices that the pressure at first drops fairly rapidly and then reaches an almost constant value. After some time the pressure is found to drop still further, but much more slowly than in the beginning. To explain this behaviour it must be remembered that the gas removed by the pump is initially present not only in the enclosed space but also adsorbed at the walls and at the surface of the components. The free gas is exhausted fairly quickly, the removal being faster the greater the pumping speed at the exit <sup>8)</sup> and the smaller the volume. The gas adsorbed at the walls and components may now be liberated, and a pressure sets in such that the amount of gas given off is equal to that removed by the pump. A state of quasi-equilibrium arises, i.e. a quasi-stationary state of flow. The rate of release of gas from the walls diminishes very slowly and the pres-

sure likewise. The pressure prevailing in the space after the quasi-stationary state has set in is governed by the amount of gas given off per second and by the pumping speed.

Mathematically, the variation of pressure with time can be described as follows.

Let the temperature be constant and uniform over the whole system. A quantity of gas, i.e. a certain number of molecules, is then entirely determined by the product of pressure and volume. Let  $Q_A$  represent the quantity of gas of kind  $A$  which is liberated from its bound state per second and  $Q_B$  that of gas of kind  $B$ , etc. The quantity of gas  $A$  with which the pump can deal per second can be described as  $S_A p_A$ , where  $S_A$  is the pumping speed for gas  $A$  and  $p_A$  the partial pressure of gas  $A$ .

If the pump is now connected directly — i.e. using no manifold — to the vessel to be evacuated, then the pressure at the pump mouth is the same as that in the space. The partial pressure  $p_A$  will then obey the relation:

$$-V \frac{dp_A}{dt} - S_A p_A + Q_A = 0, \quad \dots \quad (1)$$

where  $V$  is the volume of the vessel and  $t$  the time. For gases  $B$ ,  $C$  and so on, corresponding relations hold. If  $Q_A$  changes but slowly with time, and if  $S_A$  may be regarded as independent of  $p_A$ , the solution of this equation is:

$$p_A = p_{A0} e^{-\frac{S_A}{V}t} + \frac{Q_A}{S_A} \left(1 - e^{-\frac{S_A}{V}t}\right), \quad \dots \quad (2)$$

where  $p_{A0}$  is the value of  $p_A$  at the time  $t = 0$ . As long as  $t$  is small, i.e. at the beginning of the pumping process, then the first term on the right-hand side is considerably larger than the second, and hence almost entirely governs the value of  $p_A$ . The second term indicates the value which the pressure ultimately reaches. Naturally, the time at which the two terms are equal depends on the values of  $Q_A$ ,  $S_A$ ,  $p_{A0}$  and  $V$ . Corresponding equations again hold for the other gases ( $B$ ,  $C$ , etc.), and the total pressure, when there is no interaction between the gases, is given by:

$$p = p_A + p_B + p_C + \dots \quad (3)$$

Interaction between the gases does not take place, at least not in the evacuated space, since the mean free path at the low pressures we are considering is very long and the number of collisions between the molecules is very small compared with the number of collisions against the walls.

As can be derived directly from equations (2) and (3), the value of  $p$  ultimately established in the space is governed by the quantity of gas liberated,  $Q$ , and the pumping speed  $S$ .

In order to lower the pressure as far as possible it is necessary on the one hand to reduce to a minimum the release of gas in the space, and on the other hand to step up the pumping speed. Disregarding leaks, the release of gas from the walls can be minimized by prolonged baking out at a suitably high temperature and low pressure. Naturally, in a high-vacuum apparatus materials must be avoided that cannot readily be de-gassed, for which reason the materials used are at present limited to metals and glass.

<sup>8)</sup> The pumping speed is the ratio between the quantity of a given gas evacuated per second and the pressure of that gas (measured at the position at which the pumping speed is to be calculated). The pumping speed is usually measured in litres per second. At the inlet aperture of a pump this ratio is constant within a wide range of pressures. Where a mixture of gases is concerned, the above is still valid provided the calculation is made for each component on the basis of its partial pressure.



It cannot be concluded from the above that the pressure in a space in which the release of gas has been reduced to zero can go on falling indefinitely. The pumping speed — we shall assume for a moment that only one type of gas is concerned — does not remain indefinitely constant, but decreases when the pressure has dropped below a certain value, and finally becomes zero. The pressure at which this happens we shall call the limiting pressure for a particular type of gas. The value of this limiting pressure differs from one pump to another. Where a mixture of gases is concerned, as is always the case in practice, the limiting pressure for all components is not reached simultaneously. As soon as the limiting pressure for one of the components is reached, the composition of the gas at that moment begins to change, in the sense that it becomes richer in this component, and so on.

In the light of the above considerations we shall now examine the ionization gauge used as a pump. Its particularly favourable properties are: 1) small wall area and the fact that it can be readily out-gassed, enabling the release of gas to be reduced, and 2) the property of still being able to sorb gas even at extremely low pressures. The pumping speed, however, is not so favourable. Alpert<sup>5)</sup> estimates the pumping speed of his arrangement in the case of chemically inert gases — i.e. gases not taken up by the hot cathode — at approximately 0.02 l/sec. The pumping speed for chemically active gases, such as oxygen, is substantially higher; it is the chemically inert gases that determine the lowest attainable pressure.

A circumstance that can be a difficulty in some experiments is that the ionization gauge used as a pump does not *remove* the gas from the system. There is always a possibility that it will again be released from the gauge. If, for example, one wishes to carry out an experiment in a well-defined atmosphere having a pressure of  $10^{-8}$  mm Hg, the maximum one will wish to allow for the partial pressure of foreign gases will be  $10^{-10}$  mm Hg. A high vacuum of this order is first produced by means of the Alpert system, after which the required gas is allowed to flow in from a reservoir at such a rate that, with the ionization gauge pumping the pressure reaches the prescribed value. Proceeding in this way one is still not certain, however, that the composition of the gas will be exactly as hoped for. Since the gas-collecting components of the gauge are subjected to much more intensive ion bombardment after the filler gas is introduced, other gases, earlier taken up, can again be released and thus entirely alter the composition of the gas. This is a well-known phe-

nomenon with the Penning manometer. Of course, one can avoid this effect by switching off the manometer before admitting the gas. In that case, however, the pressure can no longer be measured, and moreover pumping ceases from the moment of switching off.

Another difficulty that can arise is concerned with the diffusion of helium from the atmosphere through the glass envelope of the apparatus. Alpert has very neatly demonstrated that this phenomenon does in fact occur and in some cases governs the lowest obtainable pressure<sup>5)</sup>. True, the effect of this diffusion can be limited by an appropriate choice of glass, but when the experiment calls for high wall temperatures, this diffusion can indeed be troublesome. The simplest method of removing this helium is to use a pump having a much greater pumping speed than is possible with an ionization gauge.

A higher pumping speed is also desirable when experiments are to be carried out on surfaces in a very high vacuum without it being possible to minimize the release of gas from the material under investigation. The only way to produce the requisite low pressure in such a case is to use a pump having a high pumping speed.

These considerations led us to set up a pumping arrangement capable of permanently removing the gas from the system and with which relatively high pumping speeds can be achieved, substantially higher than with the ionization gauge. With this arrangement we have achieved pressures which even the improved ionization gauge was unable to measure. It is estimated that the pressure in our experiments was lower than  $10^{-12}$  mm Hg.

## The pumping system

### *Choice and construction of the vacuum pump and accessories*

If the object is to build a pumping system having a high pumping speed for all gases, the use of a diffusion pump is the obvious solution. Although the speed of this pump generally decreases with increasing molecular weight of the pumped gas, this effect is not very serious<sup>9)</sup>. Diffusion pumps of high pumping speed are easy to make. Another question, however, is whether the pressure at which the pumping speed drops to zero — the earlier mentioned

<sup>9)</sup> The pumping speed varies from one gas to another approximately in proportion to the root of the molecular weight. It should also be recalled that the pumping speed depends on the pressure, so that one cannot state an unqualified value for a given gas or pump.



limiting pressure — is adequately low with the diffusion pump. The foregoing has made clear that this limiting pressure need certainly not lie at  $10^{-7}$  mm Hg, as was formerly believed because of misapprehension regarding the residual reading of the ionization gauge; it may indeed be much lower.

The principle of operation of the diffusion pump was described some years ago in an article in this journal<sup>10</sup>). For the purposes of the present article a few points may be recapitulated to clarify the problem with which we are concerned. The operation of this type of pump is based, as the name indicates, on the diffusion of the pumped gas in a stream of vapour. The stream of vapour is produced in the pump by the boiling of a suitable fluid (mercury, oil). In the following we shall speak of gas and vapour in order to distinguish clearly between the two media. The density of the vapour is much greater than that of the pumped gas, and the gas molecules are entrained in the vapour. The vapour is next condensed and the gas, which now has a greater density than at the pump mouth, then passes through a second diffusion stage, where the same process is repeated. One or more other stages may follow, and finally the gas leaves the system via a (mechanical) backing pump. In some cases the diffusion pump contains only one stage. The gas leaves this pump with a lower pressure than in the case of a multi-stage pump; in such a case more stringent requirements are imposed on the backing pump as regards the pumping speed at low pressures.

We shall now consider the factors governing the limiting pressure at the pump mouth and the method of making this pressure as low as possible.

a) The first factor is the pressure of the saturated vapour of the pump fluid used and of the decomposition products resulting from the boiling of this fluid. By interposing liquid nitrogen traps between the pump and the receiver one can greatly reduce the chance of a molecule of these vapours reaching the evacuated vessel. The traps should have the highest possible conductance for the gas stream if the ultimate pumping speed in the vessel is not to be unduly reduced; but it is also necessary to minimize the chance of a vapour molecule passing through a trap. It is not the case, however, that all decomposition products at the temperature of the trap ( $-196^{\circ}\text{C}$ ) have such a low vapour pressure as to be harmless. We have confirmed this by a number of qualitative experi-

ments<sup>11</sup>). Measurements with a mass spectrometer have shown that hydrocarbons of low molecular weight arise during the boiling of organic fluids in a pump<sup>12</sup>). For these reasons we decided on mercury as the pump fluid.

b) The second factor is the release of gas from the pump itself — particularly from the part through which the gas passes before entering the diffusion zone — and the release of gas from all components of the system to be pumped. As already mentioned, the release of gas from the components can be reduced by heating the components at low pressure and a sufficiently high temperature. With this in mind we designed our pumping system such that the most critical part of the pump — that which provides the pumping action, in our case the upper section — can be baked out in a furnace. The same applies to the traps.

c) The third factor is the back-diffusion of the gas in the pump against the vapour stream. The effect of back-diffusion can be reduced by minimizing the pressure at the pump outlet. This can easily be done by using a second diffusion pump as backing pump. In our arrangement this second pump can reach a pressure at its mouth of  $10^{-7}$  mm Hg. This also favours the out-gassing of the first pump.

If other effects are negligible, so that the limiting pressure is determined solely by the back-diffusion (assuming, again, only one kind of gas is present) there is a constant ratio in a diffusion pump between the pressure at the pump mouth and that at the outlet. The value of this ratio naturally varies with the speed of the vapour stream and with the nature of the gas. The ratio is highest, i.e. back-diffusion is most troublesome, in the case of light gases. In our pump this ratio is smaller than  $10^{-6}$  for hydrogen, at a moderate vapour speed. With a normal vapour speed, and when pumping heavier gases, this ratio is therefore appreciably smaller.

d) Finally, there is the fact that the pump fluid itself is not entirely gas-free and cannot be made so. When the vapour condenses, a certain fraction of the pumped gas dissolves in the vapour. If the above-mentioned factors a), b) and c) have been reduced to the minimum by carefully calculated measures, it may well be that this particular effect will determine the lowest obtainable pressure. In such a case, the minimum pressure is independent of the backing-vacuum pressure over a relatively wide range<sup>13</sup>).

<sup>10</sup>) F. A. Heyn and J. J. Burgerjon, Philips tech. Rev. 14, 273-276, 1952/53.

<sup>11</sup>) The decomposition products of the following were investigated: Apiezon A and C, Octoil S and Silicone DC 703. See also: C. Hayashi, J. Phys. Soc. Japan 9, 287, 1954.

<sup>12</sup>) J. Blears, J. sci. Instr., Suppl. No. 1 (Vacuum Physics), p. 36, 1951.

<sup>13</sup>) N. A. Florescu, Vacuum 4, 30, 1954.



Fig. 2 shows a cross-section of the diffusion pump designed in these laboratories, which satisfies in particular the requirement mentioned under *b*); a photograph of the pump can be seen in fig. 3.

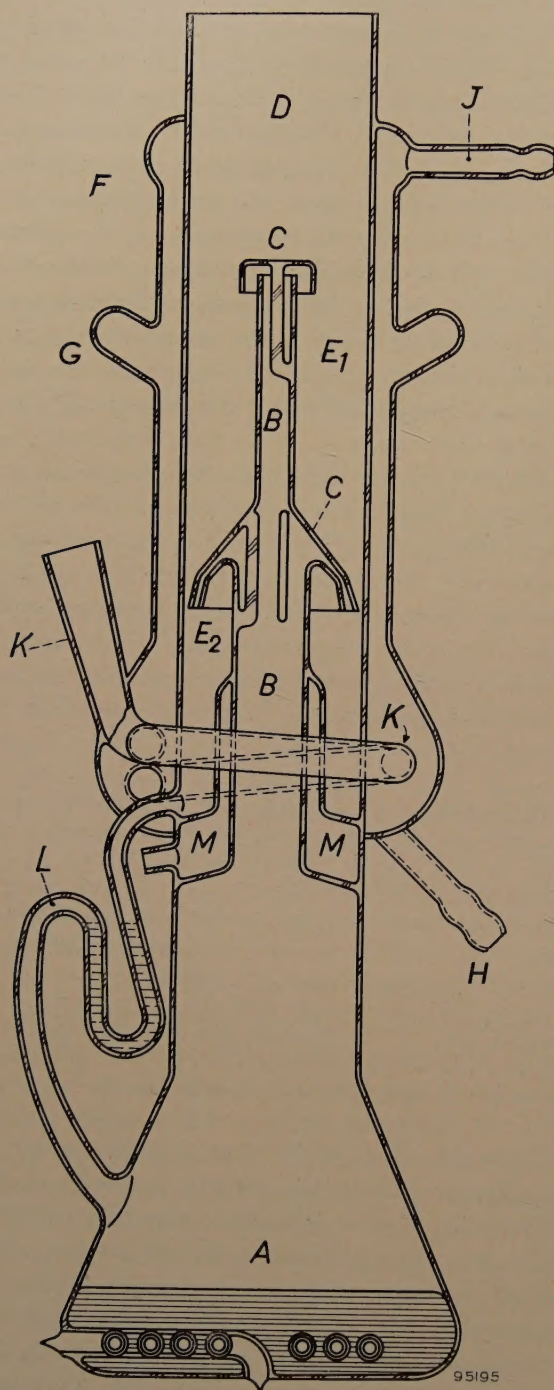


Fig. 2. Cross-section of the glass two-stage mercury diffusion pump. *A* boiler with heater. *B* funnel. *C* nozzles. *D* pump mouth. *E*<sub>1</sub> and *E*<sub>2</sub> diffusion space of the two stages. *F* water-cooled jacket with expansion bulge *G*. *H* and *J* inlet and outlet, respectively, for cooling water. *K* pump outlet; to ensure complete condensation of the mercury a fairly long section of the outlet passes through the water-cooled jacket. *L* return tube for condensed mercury; it also functions as valve and manometer. *M* space communicating with the backing-vacuum, and providing heat insulation between boiler and cooler. This insulation limits heat losses and reduces the risk of the glass breaking as a result of thermal stresses.



Fig. 3. The glass two-stage mercury diffusion pump.

It is, as the figures show, a two-stage glass pump. It is so constructed that the part responsible for the pumping action — at least as far as the first stage is concerned — can be outgassed in a furnace. One of the measures adopted to this end was to position the boiler containing the mercury as low as possible. Further particulars are given in the caption to



fig. 2. The pumping speed for nitrogen is 60 l/s over a wide pressure range; the maximum permissible backing-pressure for air, with a heater dissipation of 400 W, is 0.5 mm Hg<sup>14</sup>).

The design of the traps<sup>15</sup>) is illustrated in fig. 4. Both the inside and outside walls of the space between the inlet and outlet of the trap can be cooled — with liquid nitrogen, for example. A mercury atom is certain to strike the cold wall at least once during its passage in the trap. Two versions of the traps with different dimensions have been made having conductances for nitrogen of 25 l/s and 15 l/s; a photograph of the first type is shown in fig. 5.

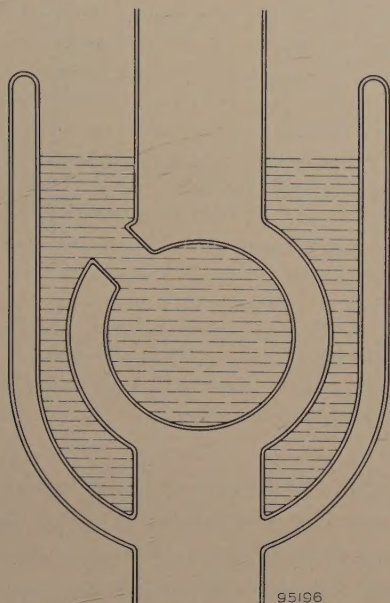


Fig. 4. Schematic cross-section of a liquid nitrogen trap having very high conductance and high condensation efficiency.

#### Layout and operation of pumping system

The design of the pumping system was based on the idea that the vessel should be outgassed longest, because at that position the pumping speed is lowest for those gases on which the traps have no effect. Fig. 6 shows a photograph of the apparatus. The glass diffusion pump passes through a hole in an asbestos-cement table, such that the upper stage of the pump is above the table top. Above the pump are, in succession, the three traps, followed

<sup>14</sup>) If the pressure difference between the inlet and outlet of a diffusion pump exceeds a certain value, the pumping speed drops sharply. The value of this reverse pressure is of the order of magnitude of 0.01 to 1 mm Hg. Owing to the use of two stages, the maximum backing pressure of the pump described here is sufficient to enable the pump to be used for "normal" high vacua in combination with merely a rotary backing-pump. For our purpose it has been found that a single-stage pump capable of being outgassed, together with a second diffusion pump, is also suitable.

<sup>15</sup>) Designed in co-operation with J. H. N. van Vucht.

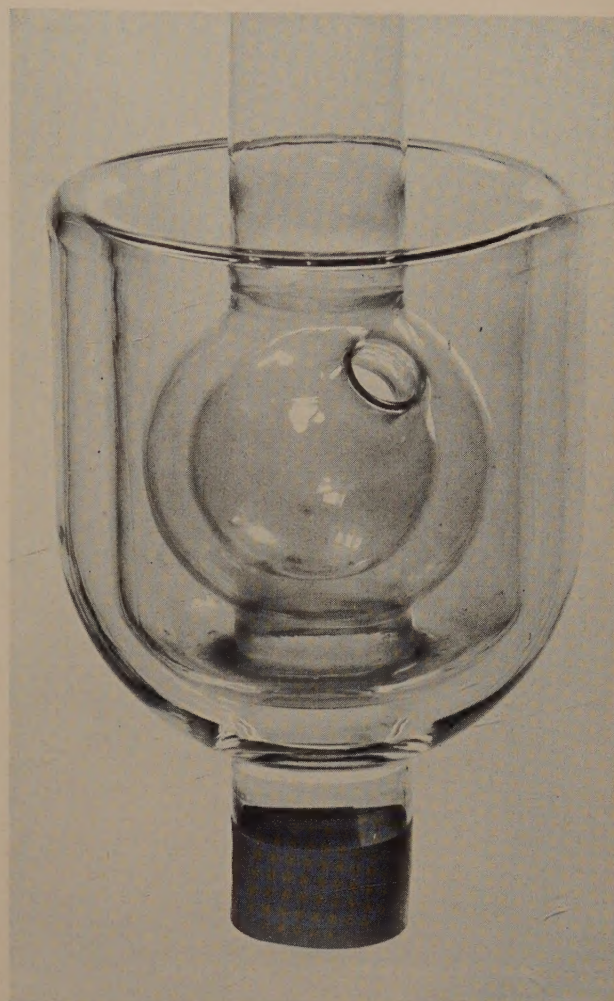


Fig. 5. One of the liquid nitrogen traps.

by the vessel and the ionization gauge. Below the table top are mounted a metal mercury-vapour diffusion pump, which acts as backing pump, and a vacuum reservoir. The reservoir is connected to the outlet of the metal pump as soon as the major part of the gas has been pumped out; up to that moment this pump is connected to a rotary-pump. The pressure in the reservoir can rise to about 5 mm Hg without seriously reducing the pumping speed of the metal pump. Also under the table top is the safety equipment responsible for switching off the electric current (both pumps, metal and glass, are electrically heated) if the cooling water pressure drops too low or if the gas pressure rises above a certain value owing to a leak. Also, in this event, a valve between the two diffusion pumps closes automatically.

The part of the installation above the table can be heated to 450 °C. The furnace in which this is done can be moved upwards after use into the position in which it appears in the photograph.



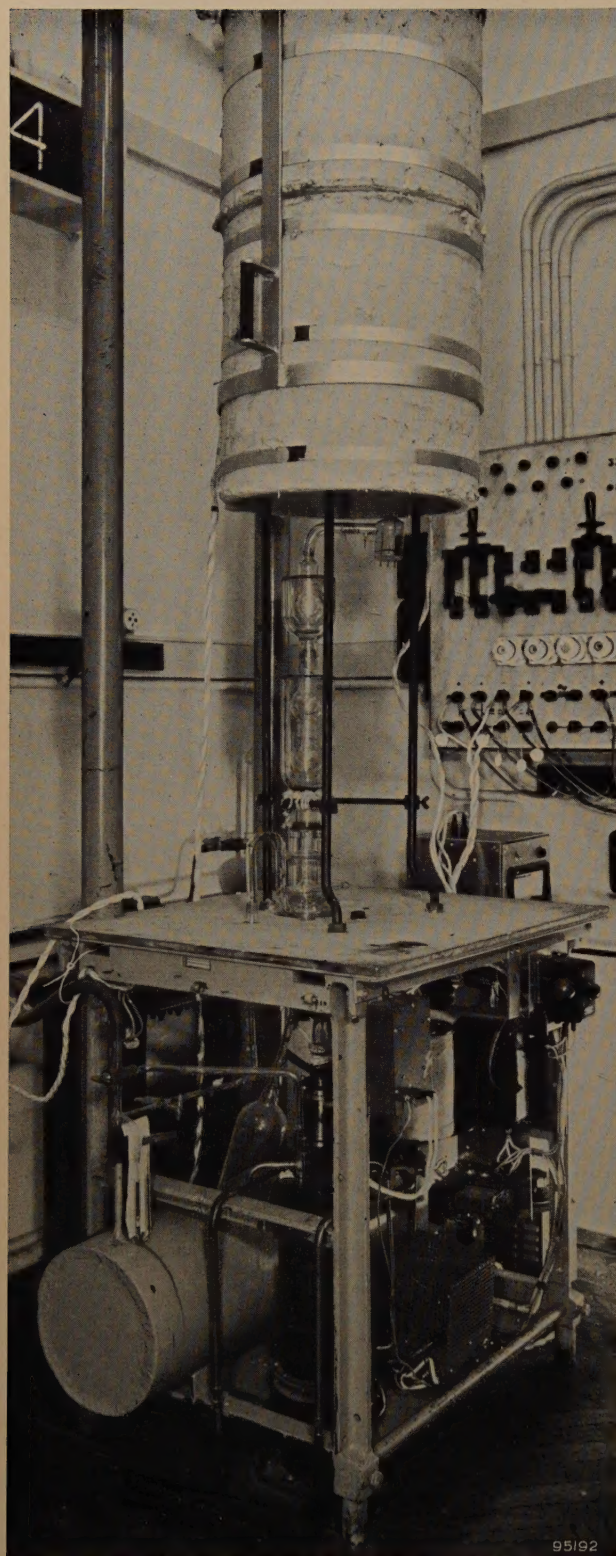


Fig. 6. The pumping system for producing ultra-high vacua. The glass two-stage mercury diffusion pump is partly above and partly below the table top of asbestos-cement. Under the table are the metal diffusion pump for the backing-vacuum, and the vacuum reservoir. Above the glass pump are three traps one above the other, a manifold bent over to the right, to be regarded as the vessel, and connected to it an ionization gauge of special design. The furnace surmounting this system can be lowered to the table top, enabling all parts above the table to be baked out.

With regard to the efficiency of the traps the following may be said. The probability of a mercury atom leaving the cooled surface on which it impinges is not zero. This probability is in fact greater here than in the case of a sealed-off space held at the temperature of liquid nitrogen; the "temperature" of the incident atom is not always equal to that of the wall after a single collision. Furthermore, the density of the mercury vapour at the inlet of the first trap is still great enough for the mean free path of the atoms to be of the order of magnitude of the dimensions of the trap. This means that the mercury atoms undergo here a by no means negligible number of mutual collisions, which enable some of them to pass through the cooler without having come into contact with a wall. In fact, mercury is still to be found at the outlet of the first trap. For this reason it is necessary to have a second trap in series with the first; if the system is to be in operation longer than about 24 hours, a third trap is necessary in order to maintain the pressure in the region of  $10^{-11}$  mm Hg.

Accordingly, measures must also be taken to keep the liquid level in the traps automatically constant. For this purpose we used a device described by Feld and Klein<sup>16)</sup> and illustrated in fig. 7. The trap *R* (here shown only schematically) is filled from a reservoir *C* by means of a syphon *S*. The pressure above the boiling fluid in *C* is regulated by the temperature in an air-filled vessel *T*. If this temperature rises above a certain value, the pressure in *T* becomes so high as to cause the mercury in a valve *V* to close an opening *O*, causing the pressure in *C* to rise. Fluid then flows through the syphon, the liquid level in the trap rises and the temperature in *T* drops. The valve opens again, the pressure above the fluid in *C* falls and the supply of fluid ceases. This system works very reliably and is simple in design.

The vacuum system is outgassed and set in operation in the following way. Initially the furnace rests on the table top. Everything above the table top is baked out and while this is happening neither the glass pump nor the coolers can function. The system is exhausted with the metal vacuum pump. After this has been in progress for some considerable time, e.g. all night, the furnace is moved up high enough to allow the glass pump to be put into operation. The remainder above the pump is kept at 450 °C. The outgassing process can now be effectively continued owing to the fact that the pressure of all gases in the system (except the mercury vapour) drops sharply as a result of the glass pump coming into operation, the pumping speed now being increased by a factor of 50. After some hours the furnace is moved up still higher to expose the first trap, which is then filled with liquid nitrogen. At intervals of a few hours the other traps are also taken into use.

The components of any experimental object that may be in the vessel are now thoroughly outgassed by heating them to an appreciably higher temperature than the wall of the vessel, which is

<sup>16)</sup> M. Feld and F. S. Klein, *J. sci. Instr.* **31**, 474, 1954.



still at 450 °C. The arrangement must of course be so designed as to make this possible. At the same time the electrodes of the ionization gauge are subjected to the same treatment. Finally, the latter section is allowed to cool. If only the ionization

to heat the remaining two traps with the vessel and the gauge for a sufficiently long period. In other respects the outgassing process is the same.

It is obvious that the vertical arrangement described here is only one example out of many possibilities. Other arrangements can be built on the same principle, possibly with more than one furnace.

It may be asked whether the increased pumping speed obtained here (as high as 5-10 l/s in the vessel) is not offset by an increase in the total release of gas from the wall, so that as regards the lowest obtainable pressure there is in fact no improvement compared with the method of pumping with the ionization gauge. In that method, it may be argued, the pumping speed was admittedly low, but at least the gas-releasing surface was small. A quantitative study of this problem (see appendix) has shown that this is not the case and that a quite substantial gain is possible in the degree of vacuum obtainable.

### The ionization gauge

In designing the ionization gauge our guiding principles were to produce a gauge having a measuring range with the lowest limit compatible with a simple and robust construction; furthermore the electrodes should be easily outgassed and the gas-release should be as low as possible. *Fig. 8* shows the design finally evolved. To comply with the first

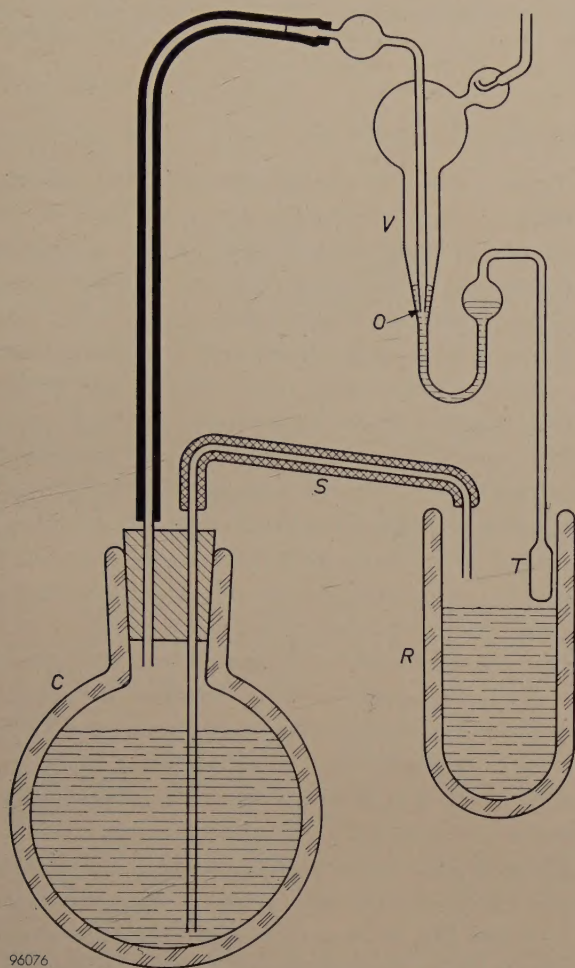


Fig. 7. Apparatus which automatically keeps the liquid nitrogen in a trap at the correct level. In the trap *R* is situated at the appropriate height a vessel *T*, which is air-filled and connected to a valve *V*. If the level of the liquid nitrogen in *R* falls, the temperature of *T* rises, the air pressure increases and at a certain moment the mercury in *V* closes the opening *O* as shown. The communication between the reservoir *C* and the atmosphere is now broken. The pressure above the boiling fluid in *C* rises, and after a short delay the syphon *S* comes into operation. As soon as the temperature of *T* drops low enough, owing to the rise in the nitrogen level in *R*, for *O* to open again, the syphon ceases to function.

gauge is connected above the last trap, the pressure in the former will then have fallen to below the lower limit of the measuring range of the gauge, i.e. below about  $10^{-12}$  mm Hg.

If the experimental object is of such a nature as to prohibit the presence of mercury vapour in the vessel at any stage in the operations, the glass diffusion pump and the first trap cannot be baked out in the furnace. In that event it will be necessary

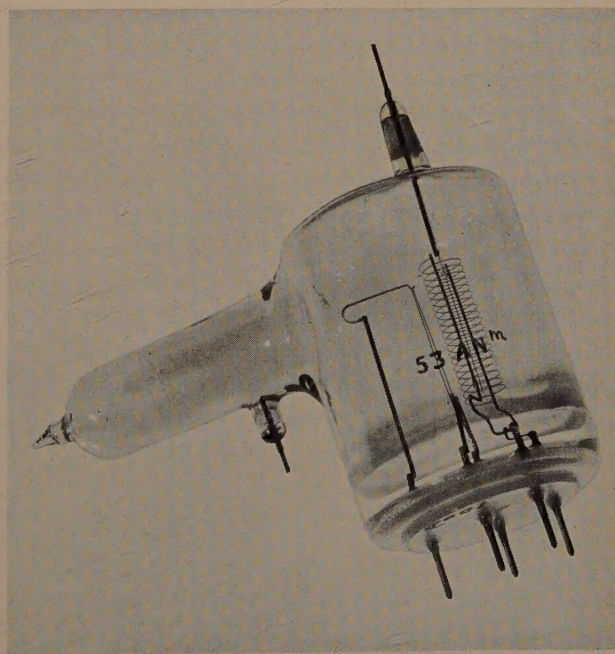


Fig. 8. The ionization gauge. The ion collector is a thin wire mounted inside the anode, the cathode being external to them. To prevent excessive heating of any one part of the wall, the cathode is mounted centrally in the glass tube. The glass wall has a conductive coating.



of the above requirements the electrode configuration is the same as in the instrument by Bayard and Alpert; to ensure minimum gas-release no stabilizer grid is used and moreover the dimensions of the electrodes have been kept fairly small. Although this entails lower sensitivity, we believe that this is more than compensated by the advantages gained. The anode is of molybdenum and consists of a cylindrical helix mounted between two wire supports. The cathode and collector are of tungsten. The power dissipated by the cathode, which is mounted centrally with respect to the glass envelope, is relatively small. Thanks to these measures no part of the tube wall undergoes any appreciable rise in temperature, and the chance of gases being released from the wall as a result of irradiation is very slight. Finally, the inside of the glass wall was made conductive. This proved to be particularly necessary for work with low electron currents. If the wall potential is not fixed it can assume all kinds of values, causing changes in the electron paths and hence altering the sensitivity. It may then happen that at the same pressure two settings of the gauge are possible, characterized by different ion currents. The wall is usually given the same potential as the ion collector<sup>17</sup>).

With the exception of the cathode, the electrodes can best be outgassed by electron bombardment. Not only the anode wires and the ion collector, but also the support wires for the anode can be heated in this way to 1100-1200 °C. A further advantage of this method of heating is that certain impurities on the electrodes, which only have a low vapour pressure even at high temperature, are dissociated under the influence of the fast electrons and rapidly disappear. The power needed to bring the anode and collector up to the required temperature is 15 to 20 W. At an anode voltage of 150 V the contribution of the X-ray effect to the collector current corresponds to a pressure of  $5 \times 10^{-11}$  mm Hg if the collector wire is 25  $\mu$  thick, and to  $2 \times 10^{-10}$  mm Hg if the wire is 150  $\mu$  thick. The upper limit of the measuring range and the sensitivity are the same as with an instrument of the older design.

The omission of the stabilizer grid calls for certain special measures in the circuitry of the instrument. If one wishes to keep the electron current constant for some time — and this is certainly very desirable during the measurement — it can

only be done by automatically regulating the temperature of the cathode, that is to say by making the electron current itself control the filament current. This of course demands additional circuitry, but this is no particular objection, especially since the electronic equipment can be used for different types of gauges (provided they do not differ unduly in their characteristics); also there is a greater chance of having to replace the gauge tube than of replacing the electronic equipment. Moreover, being less complicated in design, this gauge is cheaper than the type with stabilizer grid.

### *Electrical characteristics*

As mentioned in the introduction, the collector current  $i_c$  of an ionization gauge consists of two components, that from the positive ions originating from the gas,  $i_i$ , and that from the electrons liberated from the anode by X-rays,  $i_p$ .

These two components depend in different ways on the electron energy. The relation between  $i_i$  and the anode voltage  $V_a$  is given by a curve which greatly resembles curves of ionization probability as a function of electron energy. This curve has a flat maximum which, for most cases, lies at approximately 100 V. As regards the second component, Alpert<sup>5</sup>) has pointed out that  $i_p$  is proportional to  $V_a^a$ , the value of  $a$  lying between 1.5 and 2.

The above enables us to understand the form of the curves representing the relation between  $i_c$  and  $V_a$  shown in *fig. 9*. The various curves hold for different pressures and were obtained with two models of the new ionization gauge having collector wires of 150  $\mu$  and 25  $\mu$  thickness, respectively.

We shall first consider curves 1 and 2, which relate to the comparatively high pressure of approximately  $10^{-6}$  mm Hg. Here  $i_p$  is negligible compared with  $i_i$ , that is to say the curves show, to a good approximation, the above-mentioned relation between  $i_i$  and  $V_a$ . Turning now to curve 5, which is the lowest-lying of the curves obtained with the gauge with thick collector wire, we see that, at high  $V_a$  in particular, this curve approaches a straight line having a slope of 1.8 (dot-dash line). Evidently the X-ray limit has been reached in this case ( $i_p$  being proportional to  $V_a^a$ , as stated) and  $i_i$  is now negligible. Curve 3 occupies an intermediate position. Curve 4 holds for the same pressure, but was obtained with the other gauge; this curve, however, still exhibits the “ $i_i$  character” at this pressure. The X-ray limit of this gauge is seen to correspond with a pressure approximately ten times smaller than that which we found in the case of the gauge with the thick collector wire.

In one of our experiments the gauge with thin wire was used in the above-described pumping system for producing an ultra-high vacuum. At a

<sup>17</sup>) Nottingham (1954 Vacuum Symposium Transactions, Committee on Vacuum Techniques, Boston, Mass. U.S.A. 1955, p. 76) counteracted this effect by introducing an additional electrode surrounding the existing electrode system and situated fairly close to the glass wall. This method, however, increases the quantity of material and also makes the bake-out rather more complicated.



certain moment in the period during which the pressure was still falling slowly, measurements were carried out, the results of which are embodied in curve 6. The pressure had then already fallen to about  $2.5 \times 10^{-11}$  mm Hg. The measurements carried out when  $i_c$  was no longer decreasing are represented by curve 7. This can no longer be dis-

tinguished from a straight line corresponding to the X-ray effect; in other words the pressure — which can no longer be derived with any accuracy from these measurements — must have been lower than  $10^{-12}$  mm Hg<sup>18</sup>).

The ion current  $i_i$ , from which ultimately the pressure in the gauge must be derived, is proportional to this pressure and also, of course to the number of electrons emitted by the cathode per unit time, that is to say to the electron current  $i_e$ . We may write

$$i_i = k p i_e.$$

The proportionality factor  $k$  is dependent on the nature of the gas. This is due to the fact that the chance of ionization being caused by an electron of a certain kinetic energy differs from gas to gas. The factor  $k$  is called the sensitivity of the gauge. The sensitivity of this gauge for nitrogen, where  $p$  is expressed in mm Hg and where  $V_a$  is 150 V, is  $k = 11.2$  with the thick collector wire and 7.2 with the other.

This constant is determined in the pressure range from  $10^{-3}$  to  $10^{-5}$  mm Hg by comparing the readings of the ionization gauge with those on a McLeod gauge. In this range  $i_p$  is of such subordinate significance that the collector current  $i_c$  may be taken as equal to  $i_i$ . Alpert<sup>5</sup>) has shown that the value of  $k$  found with the type of pressure gauge described here, in this range of pressures, also holds at substantially lower pressures.

We may summarize as follows the advances made in the production and the measurement of ultra high vacua and the methods which have made these advances possible:

- 1) The X-ray limit of the ionization gauge has been lowered to the range of  $10^{-12}$  mm Hg by using the electrode system designed by Bayard and Alpert and a thin collector wire ( $25 \mu$ ).
- 2) Pressures less than  $10^{-12}$  mm Hg have been obtained by:
  - a) using a pump of high pumping speed, viz. a glass mercury-diffusion pump;
  - b) introducing three liquid nitrogen traps of special construction (maximizing the conductance and the chance of condensation) between pump and receiver;
  - c) minimizing the gas-releasing surface of the pump system and the material;

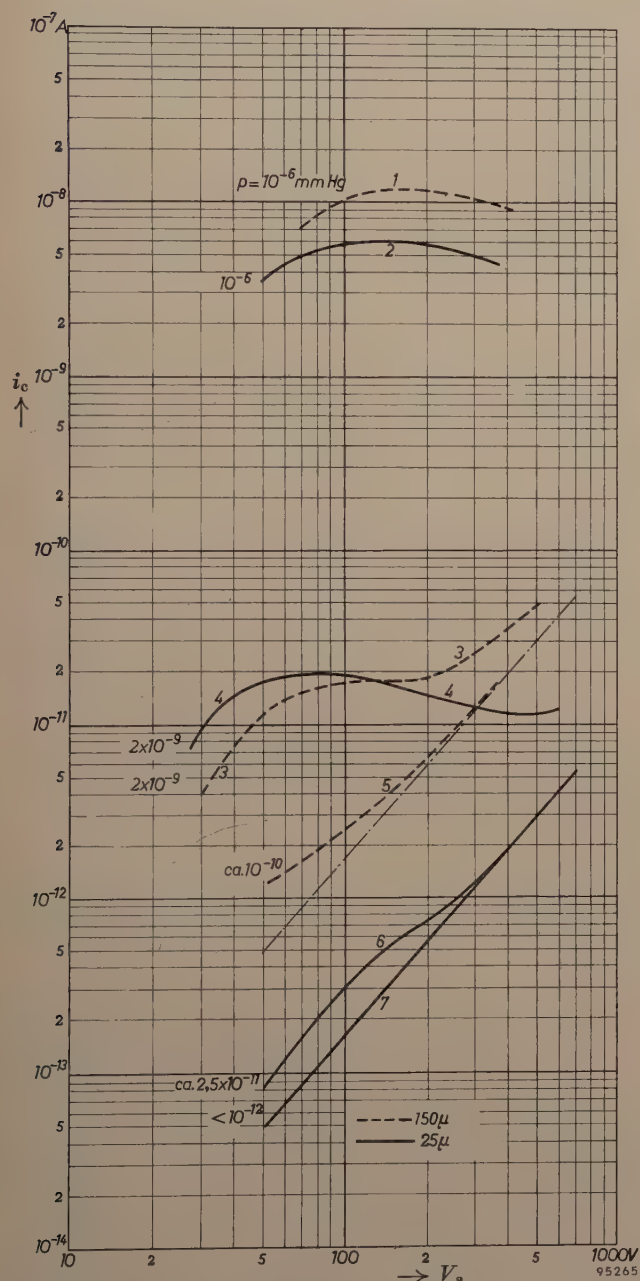


Fig. 9. The relation between the collector current  $i_c$  and the anode voltage  $V_a$  of the ionization gauge, for two versions of the gauge and at different gas pressures. The dashed curves relate to the gauge having a collector wire  $150 \mu$  thick, and the solid curves to the gauge having a collector wire  $25 \mu$  thick. At a relatively high pressure  $i_c \approx i_i$  ( $i_i$  is the ion current), and the curve has approximately the form of an ionization probability curve. At relatively low pressure  $i_c \approx i_p$ , and the curve is linear (when plotted logarithmically). The slope lies between 1.5 and 2. The measurements were made with an electron current of 1 mA. (Some of these measurements were carried out by S. Garbe and A. Klopfer of the Philips laboratory in Aachen.)

<sup>18</sup>) This pressure was calculated on the assumption that the gas concerned was nitrogen. The true composition was unknown, this gas not being a residue of the original gas contained in the vessel (e.g. air) but consisting of molecules liberated from the walls.



d) designing the pumping system such that the first pump as well as the vessel, the gauge and the traps can be outgassed in a bake-out oven (in order to produce a sufficiently low pressure for this purpose, the first pump is not followed directly by a rotary backing pump but by a second diffusion pump);  
e) reducing the back-diffusion in the glass pump (a very low backing-vacuum is also necessary for this purpose, again calling for the use of a second diffusion pump).

If it is desired to introduce instruments in the vessel for carrying out experiments, these must be outgassed just as thoroughly as the electrodes of the ion gauge. Their construction must therefore allow of high-frequency heating, for example.

#### Appendix: Calculation of the pressure distribution in the new pumping system and of the lowest pressure attainable in the ionization gauge

We shall now derive the formula for the pressure distribution in the pumping system, and with the aid of this formula we shall calculate the lowest pressure obtainable in the ionization gauge. We shall then compare this pressure with that obtainable with an ionization gauge used as a pump. In deriving the said formula we shall disregard, for the sake of simplicity, the special form of the traps employed. We regard the space to be evacuated simply as a cylindrical tube closed at one end and connected at the other end to a pump. We further assume that the gas liberated from the walls is not taken up by the coolers. (This gas may be helium, diffusing from outside through the glass wall, or gas from another source dissolved in the glass. Gases that are indeed retained in the cooler — meaning that their saturation vapour pressure at a temperature of  $-190^\circ\text{C}$  is smaller than, say,  $10^{-12}$  mm Hg — can be left out of consideration; the pumping speed for these gases is extremely high.)

The tube schematizing the system is sketched in fig. 10. The pump is connected at  $O$ ; the pumping speed is  $S$ . The radius of the tube is  $r$  and the length  $l$ . The  $x$  axis is the long axis of the tube with its origin at  $O$ . Our calculation refers to

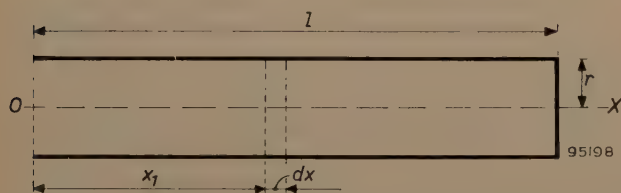


Fig. 10. Calculation of the pressure as a function of position  $x$  in a tube whose walls give off gas. One end of the tube is sealed and the other (at  $O$ ) is connected to a pump.

only one kind of gas. As soon as the quasi-stationary state has set in, the pressure is entirely governed by the gas production. The gas production per unit time and per unit area we shall denote as  $q$ .

We consider an annular element of the cylinder wall at a distance  $x_1$  from the pump and having a width  $dx$ . The gas production per unit time of this surface element is  $2\pi r q dx$ . The gas flows partly to the right and partly to the left. In the flow to the right no "loss" occurs, so that the contribution

which the element makes to the pressure at all points of higher  $x$  value is constant. Towards the left, however, the contribution decreases the shorter the distance is to  $O$ , this being due to the pumping action. In the state of quasi-equilibrium the pump removes just as much gas as the wall produces, that is to say, the contribution from the surface element mentioned to the pressure  $p_0$  at point  $O$  is

$$dp_0 = \frac{2\pi r q}{S} dx. \quad (I)$$

The gas conductance of a tube of length  $x$  and radius  $r$  is given, to a good approximation, by the formula:

$$F = \frac{1}{4} \pi r^2 v_a \frac{8r}{3x}, \quad (II)$$

where  $v_a$  is the arithmetic mean velocity of the molecules. The formula is better applicable the smaller is  $r/x$ .

When a quantity of gas  $2\pi r q dx$  flows through this tube, a pressure difference arises at the ends equal to:

$$d(p - p_0) = \frac{2\pi r q}{F} dx = 2\pi r q \frac{3x}{4\pi r^2 v_a} dx = 2\pi r q \frac{3x}{2\pi r^3 v_a} dx. \quad (III)$$

We shall now try to find the pressure prevailing at position  $x = x_1$  by calculating the contributions successively of a surface element for which  $x < x_1$ , one for which  $x > x_1$ , and of the end surface; we denote these contributions  $p_1$ ,  $p_2$  and  $p_3$  respectively.

The contribution to the pressure at position  $x_1$  in the case of all surface elements for which  $x < x_1$  is given by

$$dp_1 = 2\pi r q \left( \frac{1}{S} + \frac{3x}{2\pi r^3 v_a} \right) dx. \quad (IV)$$

This is obtained by adding equations (I) and (III). The total contribution from these elements together is thus

$$p_1 = 2\pi r q \int_0^{x_1} \left( \frac{1}{S} + \frac{3x}{2\pi r^3 v_a} \right) dx = q \left\{ \frac{2\pi}{S} r x_1 + \frac{3}{2v_a} \frac{x_1^2}{r^2} \right\}. \quad (V)$$

The contribution to the pressure at position  $x = x_1$  made by elements in which  $x > x_1$ , is

$$p_2 = 2\pi r q \int_{x_1}^l \left( \frac{1}{S} + \frac{3x}{2\pi r^3 v_a} \right) dx = q \left\{ \frac{2\pi}{S} r (l - x_1) + \frac{3}{v_a} \frac{x_1(l - x_1)}{r^2} \right\}. \quad (VI)$$

The contribution from the end surface is

$$p_3 = \pi r^2 q \left( \frac{1}{S} + \frac{3x_1}{2\pi r^3 v_a} \right) = q \left\{ \frac{\pi r^2}{S} + \frac{3}{2v_a} \frac{x_1}{r} \right\}. \quad (VII)$$

Hence, the pressure at position  $x_1$  is

$$p(x_1) = p_1 + p_2 + p_3 = q \left\{ \frac{2\pi r l}{S} + \frac{\pi r^2}{S} + \frac{3}{2v_a} \frac{x_1}{r} + \frac{3}{v_a} \frac{l x_1}{r^2} - \frac{3}{2v_a} \frac{x_1^2}{r^2} \right\}. \quad (VIII)$$

For  $x_1 = l$  we obtain the highest value of pressure:

$$p_l = q \left\{ \frac{1}{S} (2\pi r l + \pi r^2) + \frac{3}{2v_a} \cdot \frac{l}{r} \left( 1 + \frac{l}{r} \right) \right\}. \quad (IX)$$

<sup>19)</sup> The integrand in this formula differs from that in formula (V) only in that the quantity  $x$  between brackets is replaced by the constant  $x_1$ .



Because of the approximation used, this formula is valid only when  $l$  is several times larger than  $r$ , and is better applicable the larger is  $l/r$ .

In our case  $l$  was 60 cm and  $r$  was 2.5 cm. We shall now calculate  $p_l$  on the assumption that the gas concerned has approximately the same molecular weight as air. At the same value of  $q$ , the pressure in the case of lighter gases is lower. Substitution of the above values for the dimensions and of 60 l/s for the pumping speed yields the result:

$$p_l = 0.039 q.$$

For comparison we shall calculate the pressure that would be obtained with an ionization gauge used as a pump. We put the area of the gas-releasing surface at 300 cm<sup>2</sup>. At a pumping speed of 20 cm<sup>3</sup>/s we then find  $p = 15 q$ , which is higher by a factor of almost 400.

From formula (IX) we can ascertain the manner in which the dimensions of the pumping system can be varied without changing  $p_l$ . We must remember in this connection that  $S$  is approximately proportional to the square of the diameter of the pump. If we ensure that the ratio between pump and tube diameters remains unchanged when changing the dimensions, then  $S$  is also proportional to  $r^2$  and formula (IX) transforms into a function of  $l/r$ . In this case, therefore,  $p_l$  will not change as long as  $l/r$  is kept constant. We find, then, that  $p_l$  is approximately invariant when the linear dimensions of tube and pump are changed by the same factor.

**Summary.** The lowest pressure that can be achieved in a vessel connected to a vacuum pump is determined by the gas liberated from the walls and by the speed of the pump. After the space has been exhausted a quasistationary state of flow sets in, whereby the pump removes just as much gas as the walls produce. The pumping speed is not, however, a constant, but rapidly falls to zero when the pressure approaches a certain limiting value, which differs from one gas to another. It is found that the limiting pressure of a normal mercury-diffusion pump can be lowered to below  $10^{-12}$  mm Hg. The pressure difference between the inlet and outlet of the pump (the backing-pressure) was lower than  $10^{-6}$  mm Hg. A second diffusion pump is used to produce the backing-vacuum. Diffusion pumps, moreover, have a high pumping speed (at pressures well above the limiting pressure). With a pumping system designed in the Philips laboratory at Eindhoven a pressure of less than  $10^{-12}$  mm Hg has been obtained with the aid of a glass two-stage diffusion pump, preceded by a metal diffusion pump for producing the backing-vacuum; this system contains three liquid nitrogen traps between pump and evacuated vessel. The traps prevent the penetration of mercury vapour into the vessel. The pumping system is so designed that the vessel with attached ionization gauge, the traps and the first stage of the glass pump can be outgassed by heating them in an oven to 450 °C. The traps have a very high conductance and condensation efficiency. The pumping speed after the last trap is still 5-10 l/s. The wall area of the space to be evacuated is kept as small as possible. The ionization gauge is of the type with central ion collector (wire 25  $\mu$  thick), the design being such that the contribution of the X-ray effect to the collector current corresponds to a pressure of only  $5 \times 10^{-11}$  mm Hg. The anode and the collector can be outgassed by electron bombardment to a temperature of 1200 °C.



## AN APPARATUS FOR TESTING THE SOLDERABILITY OF WIRE

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The electrical connections between components in electric apparatus are usually made by soldered joints. These joints, particularly in mass production, must be effected quickly and reliably. To achieve this the solder must rapidly and thoroughly wet the metals to be joined. In order to investigate the conditions to this wetting process, that is to ascertain the "solderability" of the metal parts, an objective criterion is needed. To this end a soldering test is carried out under standardized conditions that closely approach practical conditions.

For the general investigation of solderability wide use is made of what is known as the "spread" test. At Philips this test is performed in the following way. A measured quantity of solder (say 0.2 g) is placed, together with a suitable amount of flux, on a square sample ( $30 \times 30 \times 0.3$  mm) of the metal to be soldered. This small plate is then heated by floating it on a bath of molten solder kept at constant temperature. When the solder on the plate melts, it wets part of the surface, the size of the patch wetted depending on the conditions. Approximately 15 seconds after beginning to melt, the solder has usually stopped spreading. The area of the patch wetted after this time is taken as a measure of the solderability under the conditions of the test<sup>1)</sup>.

The spread test is well adapted for investigating the conditions favourable for dip-tinning, a process in which metal parts are simply given a thin coating of solder by dipping them in a bath of molten solder. The test does not so readily allow conclusions to be drawn with regard to the *joining* of wires by dip soldering or by soldering-iron work, however. A drawback in this respect is that the spread time of 15 seconds is long compared with the time of about 2 seconds which is common for work with the soldering-iron. An example will make this clear. If, instead of a solution of pure rosin in alcohol, a more active flux be used, consisting of pure rosin with 10% ureum in alcohol, the solder will initially spread faster (see curves *A* and *B* in fig. 1). This very favourable effect is not revealed, however, by the results of the test. As seen from the figure, the test indicates the pure rosin to be the better of the two fluxes. The error in the conclusion is still more evident when pure rosin in alcohol is compared with

an even more active flux, namely hydrazine-hydrochloride in water (fig. 1, curve *C*). In many cases the spread test also fails to reveal the effect, generally favourable, of a higher soldering temperature. The correlation between practice and the results of such tests is therefore not very good. Attempts have

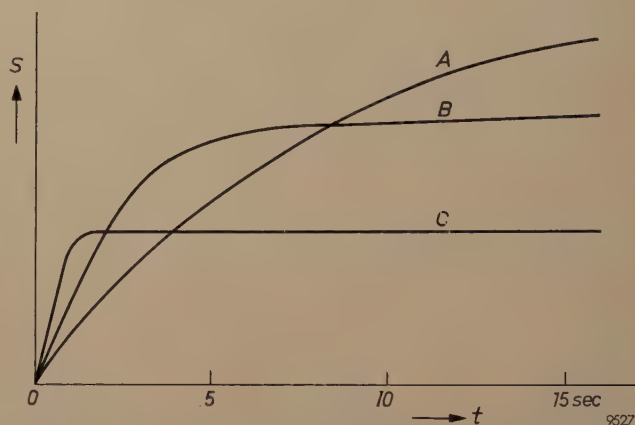


Fig. 1. The area *S* of a heated metal plate wetted by flowing solder, as a function of time in the "spread" test. The three curves relate to tests in which only the flux was varied: curve *A* refers to pure rosin in alcohol, curve *B* to pure rosin with 10% ureum in alcohol, and curve *C* to hydrazine hydrochloride in water. The area wetted after 15 sec is taken as a measure of the solderability. Interpreting the tests on this basis, the favourable effect of an initially rapid spread is lost.

been made to overcome these drawbacks by measuring the wetted area after shorter intervals, while the spreading is still in progress. Apart from photographic methods, which are rather cumbersome, all the methods hitherto proposed have been wanting in accuracy. A further disadvantage of the spread test is the somewhat lengthy procedure of determining numerically the size of the moistened area. What is more, this form of test is obviously far from ideal when the metal to be soldered is in the form of *wire*, which is usually the case in the electronics industry, where the most common components — resistors and capacitors — have wire connections.

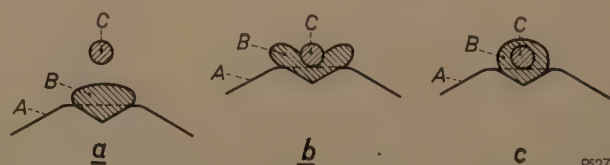


Fig. 2. Principle of the solderability test developed at Philips. *a*) In a hollow in the heated soldering block *A* lies a globule of solder *B*, above which is clamped the wire *C* under investigation. *b*) The block is raised; the wire splits the globule in two. *c*) The two halves meet at the top of the wire and flow together. The time between the splitting and the flowing-together is taken as a measure of the solderability of the wire.

<sup>1)</sup> For other versions of this test see, for example, W. R. Lewis, Notes on soldering, published by the Tin Research Institute, Greenford, Middlesex, England, 1948; also L. Pessel, Symposium on solder, Amer. Soc. Test. Mat., Special technical publication No. 189, p. 159-174, 1956.



With this in mind we have developed a method which is specially adapted for testing the solderability of wire and which avoids the drawbacks described above. The measure of solderability is taken as the time needed for a drop of solder to flow entirely around a wire of the metal under investigation. The principle of the test is illustrated in *fig. 2*. In a small hollow in a block of chromium-plated chrome-nickel steel (the soldering block) a measured quantity of the solder is introduced. The exact quantity is obtained by punching a pellet out of a bar of solder rolled to a certain thickness. The soldering block is electrically heated. When the solder has melted it remains in the hollow in the form of a globule, for solder does not wet chromium. The wire to be investigated is clamped centrally above the globule of solder, having been dipped beforehand in the flux. The soldering block is now raised so that the wire cuts the drop of solder in two. The two halves creep up round the wire at a speed which depends on the conditions. When they come into contact at the top of the wire they flow together with an abrupt movement. The instant at which this happens can be observed very precisely.

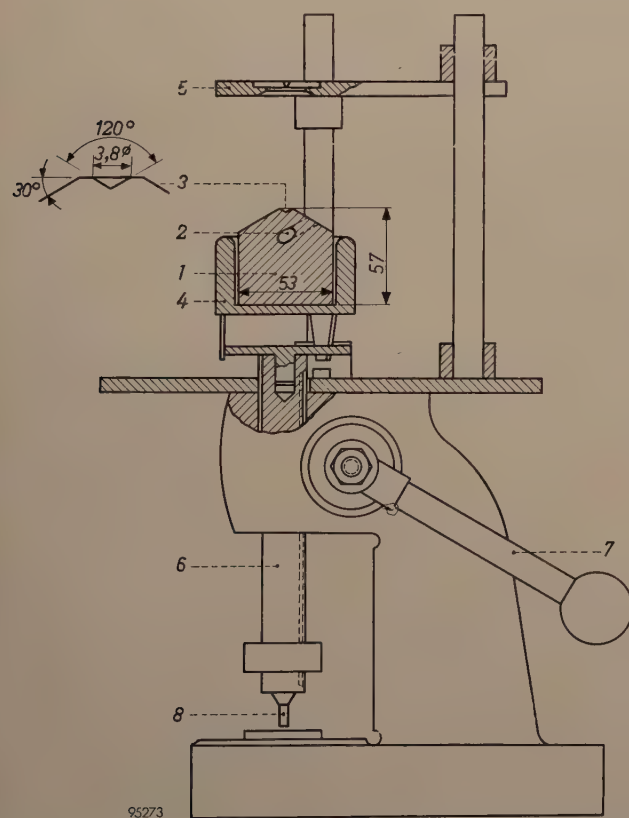


Fig. 3. Schematic cross-section of the apparatus for testing the solderability of wire. 1 soldering block, 2 hole for thermometer, 3 hollow for the drop of solder, 4 heater jacket, 5 wire holder, 6 toothed plunger, 7 lever for raising the soldering block, 8 tool for punching out the solder pellets. The shape and dimensions (mm) of the hollow for the drop of solder are given in the inset sketch.



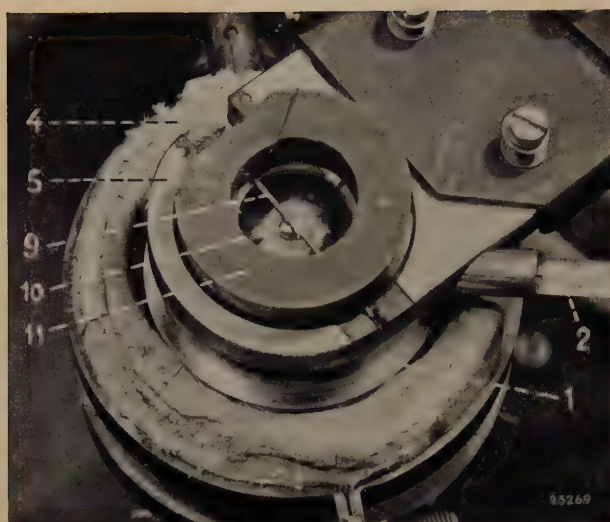
Fig. 4. Photograph of the apparatus for testing the solderability of wire. 2 is the thermometer. Other numbers as in *fig. 3*.

The time elapsing between the splitting of the globule and the flowing together at the top of the wire is measured with a stopwatch. The shorter this "soldering time", the better the solderability. The results show very good correlation with practice.

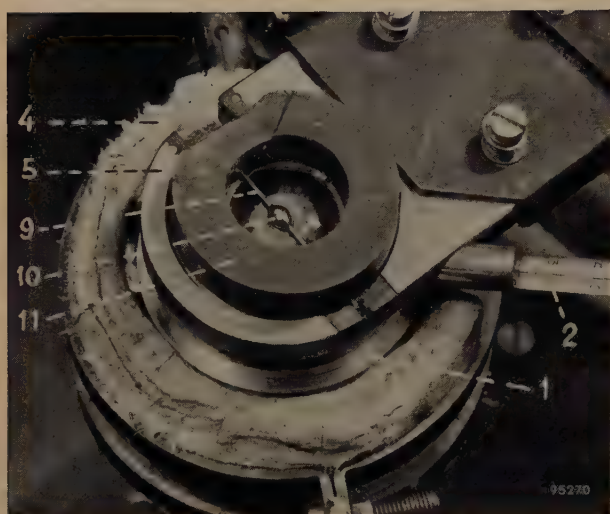
The temperature of the globule is measured with a mercury thermometer, inserted in a cavity in the soldering block. The correction to be made for the thermometer reading is ascertained by replacing the solder on the block by salts of known melting point. The construction of the apparatus is illustrated schematically in *fig. 3* and a photograph is shown in *fig. 4*. In *fig. 5a* and *b* can be seen the molten drop of solder immediately before and after the instant it flowed around the wire.

In order that the results obtained with each apparatus should be comparable it is necessary to ensure that they are identical in certain essential points. This applies, for example, to the dimensions of the hollow that holds the globule of solder, for they determine the height of the globule, and a higher globule (for example) will flow more rapidly around the wire (*fig. 6a*). The dimensions of the hollow are given in *fig. 3*. With these dimensions and with a solder globule of 190 mg, the apparatus is suitable for testing wire of 0.5-1.0 mm diameter. The other dimensions of the soldering block and the material of which it is made influence the supply of





a



b

Fig. 5. Close-up views of a globule of solder, a) split by the wire under test and b) after flowing around the wire. 2 thermometer, 9 test wire, 10 globule of solder, 11 ferroxidure magnet for clamping the wire. Other numbers as in fig. 3.

heat to the globule, whilst the length of the wire and the way in which it is clamped affect the dissipation of heat. In these matters too, therefore, individual instruments must be closely identical.

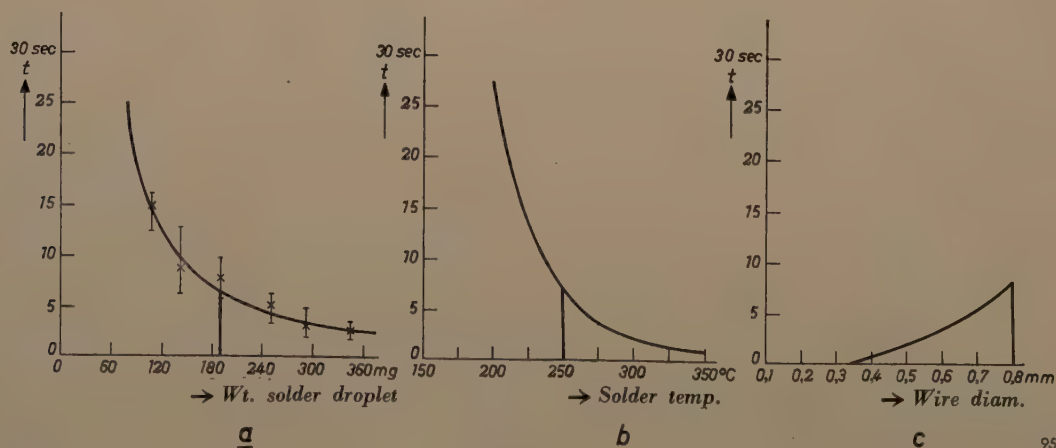
With the test method described, an investigation has been carried out into the influence of various factors on the soldering process, the principal factors being:

- 1) composition and quantity of the solder;
- 2) composition and quantity of the flux;
- 3) composition and surface state of the metal to be soldered;
- 4) temperature.

The investigation was prompted in particular by the need for greater control of the dip-soldering process as used for printed wiring<sup>2)</sup>. In this process all connections between the mounted components and the printed wiring are simultaneously soldered by dipping in a bath of molten solder. In hand soldering, with a soldering-iron, any difficulty in soldering particular wires can usually be overcome by suitable raising the soldering temperature or by applying a more active flux. In the dip-soldering of printed wiring the difficulties are much greater: often the base cannot be subjected to too high a temperature without the printed wiring becoming loose, and there is a considerable risk that a highly active flux may creep and create leakage paths. These considerations impose more stringent requirements on the solderability of the connection wires.

The investigation was designed round certain fixed conditions, one of the variables being changed at a time for each experiment. The fixed conditions were chosen with a view to obtaining a soldering

<sup>2)</sup> See for example R. van Beek and W. W. Boelens, Printed wiring in radio sets, Philips tech. Rev. 20, 113-121, 1958/59 (No. 5).



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Fig. 6. Influence of (a) quantity of solder, (b) temperature and (c) diameter of wire, on the solderability of the wire. Soldering time  $t$  is plotted vertically. The points ( $\times$ ) plotted in (a) are the averages for 10 experiments; the spread in the observations is also shown.



time that was not so short as to make the influence of favourable factors scarcely perceptible. These conditions were:

Solder: 60% tin, 40% lead; globule quantity 190 mg (22.4 mm<sup>3</sup>).

Flux: 40 parts by weight of pure rosin and 60 parts by weight of alcohol, applied to the wire by dipping.

Wire: copper wire of 0.8 mm diameter; degreased successively in trichlorethylene and acetone.

Temperature: 250 °C.

The average "soldering time" under these conditions was about 7 sec.

Very favourable conditions yielded times shorter than 1 second. The effects of variations in the quantity of solder, the temperature and the wire diameter can be seen in fig. 6a, b and c. There is a fairly wide spread in the results, but the averages of ten tests for each measuring point lie reasonably well on the curves. Since each measurement takes little time, it is practicable to make a number of repeat observations under the same conditions.

A slight change in the composition of the solder can cause a marked change in the soldering time. The addition of 1% cadmium or zinc lengthens the soldering time to more than 30 sec. On the other hand, the addition of 1% silver has a slight shortening effect. With solder consisting of pure tin the soldering time is longer than 30 sec.

The experiments on the influence of the solder composition were repeated at a soldering temperature of 300 °C. The soldering times were found to be fairly short and the differences less marked. With printed wiring, however, low-temperature soldering is necessary and in that case the composition of the solder is of importance.

As regards the composition of the flux it was found that the concentration of the pure rosin solution must be at least about 25% if it is to be applied by dipping the wire in it. With rosin concentrations

of 20% the soldering time was frequently in excess of 30 sec. Increasing the content from 25% to 40% gives hardly any further improvement. The addition of a quantity of ureum equal to 10% of the quantity by weight of pure rosin reduces the soldering times to less than 1 sec. The same short times are obtained with solutions of the monohydrochloride or dihydrochloride and monohydrobromide or dihydrobromide of hydrazine in water. Owing to their rapid decomposition, the concentration of these four compounds has a definite lower limit. The limiting concentration for the first three lies at about 3% and for the fourth at about 2.5%; at these concentrations soldering times shorter than 0.1 sec as well as longer than 30 sec are found. The best results with zinc chloride are found for an approximately 10% solution in water, the average soldering time then being 1.2 sec.

The composition of the metal to be soldered and the condition of the metal surface are extremely important factors. It is well known that thin coatings of certain metals, e.g. tin, applied by electroplating or by hot-dipping to copper wire can greatly improve the solderability. However, moisture on the surface, and, in particular, heating of the wire (say, 2 hours at 190 °C, as for example in the stove-enamelling of painted parts) can reduce this effect and make the solderability of tinned wire a great deal poorer. Less trouble in these respects is obtained with hot-dipped coatings of tin-lead (50% Sn, 50% Pb) and electroplated layers of tin-zinc (80% Sn, 20% Zn). The spread in the results of temperature and moisture tests is often greater for hot-dipped than for electroplated coatings.

It should be noted that a soldering block of aluminium was used for the investigation described here. Because of its harder wearing properties the chromium-plated chrome-nickel steel block, already mentioned, was later adopted for routine tests.

J. A. ten DUIS.



# THE LIFE OF BALLASTS FOR GAS-DISCHARGE LAMPS

## II. CAPACITORS

by T. HEHENKAMP.

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*With the spread of lighting by gas-discharge lamps there has been an enormous increase in the use of capacitors, which are used in ballasts with the object, among other things, of improving the power factor. It has therefore become a matter of considerable economic importance to produce capacitors for this purpose which are both long-lived and low-priced. The problems involved are more complicated than in the case of transformers and chokes for lamp ballasts, but they have been so far solved that capacitors fabricated by modern methods can now be used in ballasts without misgivings.*

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Many ballasts for gas-discharge lamps contain capacitors, the chief purpose of which is to improve the power factor. These capacitors are of the type having a dielectric of impregnated paper and a hermetically sealed casing. The dielectric is impregnated either with a mineral oil (sometimes vaseline) or with a synthetic oil such as chlorodiphenyl.

The life of these capacitors is just as important as that of the transformers or chokes described in Part I of this article <sup>1)</sup>. The primary factor governing the life of the latter is the temperature of the insulating material. The life of capacitors, however, is a problem of much greater complexity, for it is influenced not only by insulation temperature but also by the field strength. This is substantially higher than in the transformers and can be as much as 100 to 200 kV/cm (10 to 20 V/ $\mu$ ) <sup>2)</sup>. The reason for the use of such high field strengths is that the volume of a capacitor is about inversely proportional to the square of the field strength, so that raising the field strength allows an appreciable reduction of dimensions and price. At field strengths of this order, however, ionization is quite likely to occur in holes in the dielectric. This damages the dielectric material and leads to breakdown.

The influence of temperature on the life of the dielectric in a capacitor is comparable with that found with transformers (see Part I). Although the temperature-rise due to dielectric losses is not considerable, it must be remembered that the capacitors often operate in a high ambient temperature (owing to the proximity of warm transformers and lamps) and that they cannot withstand nearly such

high temperatures as transformers. One of the reasons for the latter fact is that the dielectric losses rise steeply with temperature, causing the capacitor to enter into a state of thermal instability <sup>3)</sup>.

### Thermal stability

If one determines the loss  $P_1$  in a capacitor at constant field strength and varying dielectric temperatures, one finds a curve that rises steeply when a certain temperature region is reached (1 in fig. 1). The dissipation of heat  $P_2$  to the surrounding medium is almost proportional to the dielectric's mean rise in temperature  $\Delta T$  above the ambient temperature:

$$P_2 = K \Delta T.$$

The proportionality constant  $K$  depends on certain constructional parameters (such as internal thermal conductivity and surface heat transfer coefficient) and on the external cooling conditions. The internal thermal conductivity causes a temperature gradient in the dielectric, making the temperature in the middle of the capacitor higher than at the walls. In ballast capacitors, however, this temperature gradient is not important and can be disregarded in approximate calculations.

Assuming the capacitor to be continuously loaded at an ambient temperature  $T_0$  we can determine approximately the temperature acquired by the dielectric in the steady state. Through point  $T_0$  on the abscissa of fig. 1 a straight line 2 is drawn which makes an angle  $\alpha$  with this axis such that

$$\tan \alpha = \frac{P_2}{\Delta T} = K.$$

<sup>1)</sup> T. Hehenkamp, The life of ballasts for gas-discharge lamps, I. Transformers and chokes, Philips tech. Rev. 20, 59-68, 1958/59 (No. 2/3).

<sup>2)</sup> The numerical values of field strengths and voltages throughout this article refer to the r.m.s. values.

<sup>3)</sup> J. Coquillion, Condensateurs au papier imprégné de pyralène pour le courant alternatif, Electricité 40, 237-243, 1956.



In general this line cuts the loss curve  $l$  at two points. It can be seen that the point of intersection on the right represents an unstable state of equilibrium, while the point on the left is a stable state. As the temperature of the dielectric rises, it will thus settle down to the value  $T_d$  corresponding to the left point of intersection.

If the ambient temperature be raised, the straight line will move, maintaining the same slope, to the right. The two points of intersection approach each other and finally coincide to form a single point of contact. In this state

$$\frac{dP_1}{dT} = K.$$

This is the *stability limit*, and it is reached at an ambient temperature  $T_s$ . Thermal stability is no longer possible at an ambient temperature higher than this; the capacitor then goes on rising in temperature until it breaks down. Breakdown occurs at, or close to, the hottest part of the dielectric, i.e. in the centre. By switching off the capacitor a few moments before it breaks down and unwinding the paper, one can find the place where the breakdown would have occurred as a darkly discoloured patch caused by overheating (fig. 2).

A capacitor of the same type subjected to a different electrical load gives a loss curve differing from  $l$  in fig. 1, and hence has a different critical

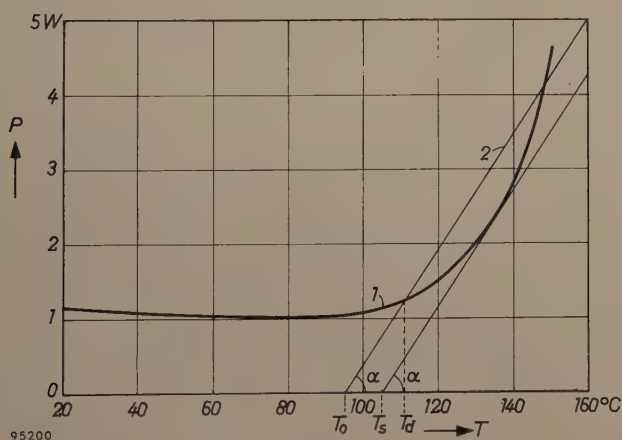


Fig. 1. Curve 1: Loss  $P_1$  in a 3.8  $\mu\text{F}$  capacitor as a function of the temperature  $T$  of the dielectric, at constant field strength ( $22 \text{ V}/\mu^2$ ) and constant frequency (50 c/s). The dielectric consists of three layers of paper 8  $\mu$  thick, impregnated with pentachlorodiphenyl.

Curves 2: Graphical construction for finding the equilibrium temperature  $T_d$  of the dielectric for an ambient temperature  $T_0$ . Through point  $T_0$  on the abscissa a straight line 2 is drawn at an angle  $\alpha = \tan^{-1} K$ ; the line 2 intersects with curve 1 in two points; the left point of intersection corresponds to the temperature  $T_d$ . The highest ambient temperature at which a stable state can still be established is the value  $T_s$  on the abscissa, which is the point where the line 2 tangential to  $l$  intersects the  $T$  axis.



Fig. 2. Two strips of paper unwound from a capacitor which was switched off just before thermal instability would have caused breakdown. Left, a strip from the outside (coldest) layers; right, a strip from the inside (hottest) layers. The right strip has turned brown due to overheating.

ambient temperature  $T_s$ . If the field strength be lowered, for example, the losses decrease, which means, according to fig. 1, a higher value of  $T_s$ . By plotting the same graph for different field strengths one finds the relation between field strength and  $T_s$ . Fig. 3 shows the result; the line found for this relation is usually straight or slightly curved.

In fig. 3 the region right of the line is unusable: a capacitor loaded in this region would soon break down. Left of the line stability is assured, at least as long as the losses do not increase unduly owing to ageing of the dielectric.

The thermal stability limit can also be found by a direct test method. The capacitor, under the appropriate electrical load, is placed in an oven whose temperature is somewhat lower than the value of  $T_s$  estimated to correspond to the field-strength selected. Once a state of equilibrium is reached, the oven temperature is raised in small steps, with a pause after each step to allow equilibrium to be re-established. When equilibrium no longer sets in, the oven temperature is then about equal to  $T_s$  and the capacitor soon breaks down.



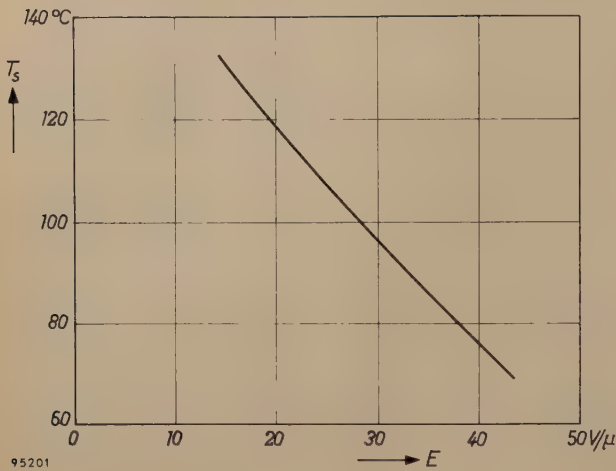


Fig. 3. The ambient temperature  $T_s$  at which the limit of stability is reached, as a function of the field strength  $E$ . The values of  $T_s$  are determined graphically as indicated in fig. 1, for capacitors having a loss curve 1 as in fig. 1.

### Life at high temperatures

When a capacitor is loaded with a certain field-strength  $E$  at an ambient temperature  $T_0$  which is but little lower than the value of  $T_s$  corresponding to  $E$ , the capacitor must obviously be expected to have a short life. The reason is that as soon as the dielectric begins to deteriorate under the influence of the electrical and thermal loading, the capacitor will become unstable, owing to the increasing losses, at a lower ambient temperature than was the case in the initial state (fig. 4). The difference  $T_s - T_0$  (fig. 1) is according to this reasoning a measure of the reserve available in the capacitor. This reserve and the rate at which it drops to zero (the aging rate) determine the life of the capacitor. This is true within a limited temperature range, as appears from life tests at high ambient temperatures.

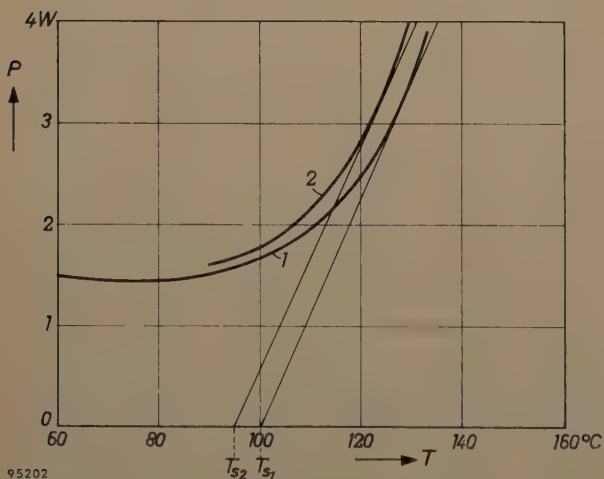


Fig. 4. Loss curve of a  $3.8 \mu\text{F}$  capacitor for a field strength of  $25 \text{ V}/\mu$  and a frequency of  $50 \text{ c/s}$ , in the initial state (curve 1) and after 250 hours loading at  $25 \text{ V}/\mu$  and  $90^\circ\text{C}$  (curve 2). Owing to the aging of the dielectric the stability limit has dropped from  $T_{s1} = 100^\circ\text{C}$  to  $T_{s2} = 95^\circ\text{C}$ .

An example is given in fig. 5, which represents the results of life tests on capacitors of  $4 \mu\text{F}$  having a  $30 \mu$  paper dielectric. When the ambient temperature is lowered the life is found to increase enormously. This suggests that at, say,  $T_0 = 40^\circ\text{C}$ , the capacitor would have an extremely long life. Such an extrapolation, however, is very dangerous, for this expectation is by no means always borne out in practice. The reason for this is the increase in the degree of ionization at low temperatures, which we shall now discuss.

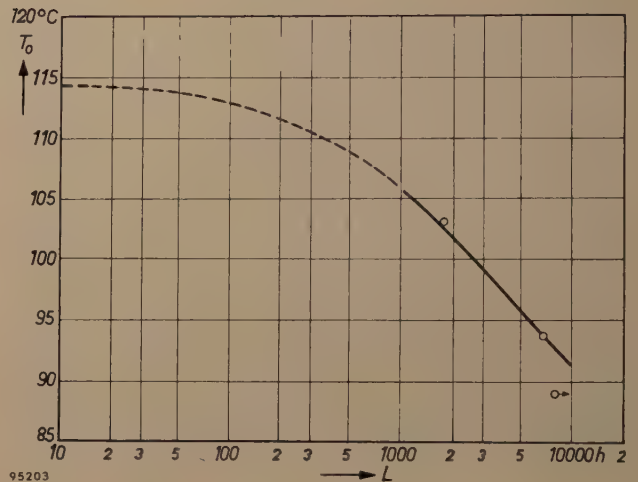


Fig. 5. Ambient temperature  $T_0$  at which capacitors having a dielectric of three layers of paper  $10 \mu$  thick, impregnated with vaseline, and under a load  $E = 17.5 \text{ V}/\mu$ , attain a life  $L$ . The stability limit  $T_s$  for  $E = 17.5 \text{ V}/\mu$  lies at  $114^\circ\text{C}$ . At this ambient temperature the capacitor has a life of only a few hours. The test at  $89^\circ\text{C}$  was still not concluded after 8000 hours.

### Ionization in the dielectric; life at low temperatures

If there is a gas bubble in the dielectric, a disproportionately large field concentration will occur over it, for the dielectric constant of the gas is only  $\frac{1}{4}$  to  $\frac{1}{6}$  of that of the impregnated paper. Since the field strength in the paper is already high, the field in the gas can easily reach a value at which ionization can occur (corona<sup>4</sup>). As a result the dielectric around the gas bubble becomes impaired (fig. 6a and b). This leads to increased losses (fig. 7), while the breakdown potential decreases<sup>5</sup>.

In the greatly simplified case of a uniform field — gas bubble with flat bounding surfaces perpendicular to the electric field and a thickness which is small in proportion to the other dimensions — the field strength at which ionization begins to occur can

<sup>4</sup> C. de Lange, Pressure condensers, Philips tech. Rev. 4, 254-259, 1939.

<sup>5</sup> T. W. Liao, Corona and its effects on insulation, Insulation, March 1957, pages 18-21.



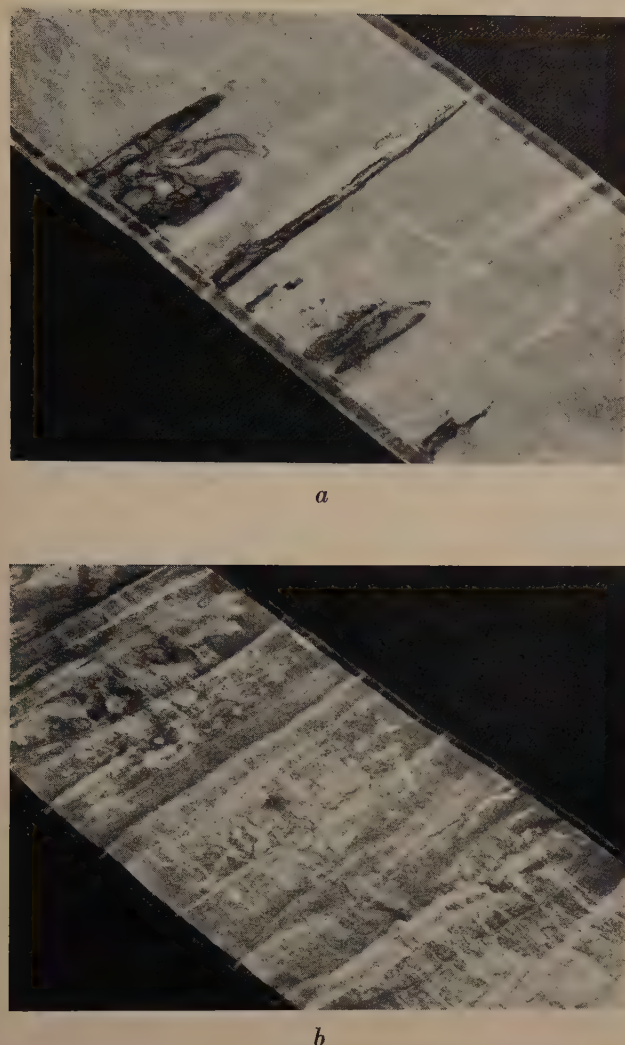


Fig. 6. Illustrating how ionization damages a dielectric impregnated (a) with pentachlorodiphenyl and (b) with vaseline. In (b) breakdown has occurred at one of the damaged regions.

be ascertained with the aid of Paschen's law, which is represented graphically for air in *fig. 8*. In reality the conditions are, of course, much more complex. Owing to the irregular structure of the paper fibres and the presence of the impregnating agent, the field strength will vary from one place to another. We may therefore expect no more than a rough correspondence to the case of a uniform field.

For this reason the potential at which ionization begins to occur must be measured in every individual case. Ionization is accompanied by slight irregularities in the current through the capacitor. The presence of ionization can therefore be demonstrated by means of a circuit which amplifies these irregularities and makes them visible on a cathode-ray oscilloscope<sup>6)</sup>. A suitable circuit for this purpose

<sup>6)</sup> R. J. Hopkins, T. R. Walters and M. E. Scoville, Development of corona measurements and their relation to the dielectric strength of capacitors, *Trans. Amer. Inst. Electr. Engrs.* **70-II**, 1643-1651, 1951.

is shown in *fig. 9*. The capacitor under investigation  $C_x$  is connected to another capacitor  $C_0$  of good quality, not heavily loaded, and two resistors to form a bridge circuit. The one diagonal is fed

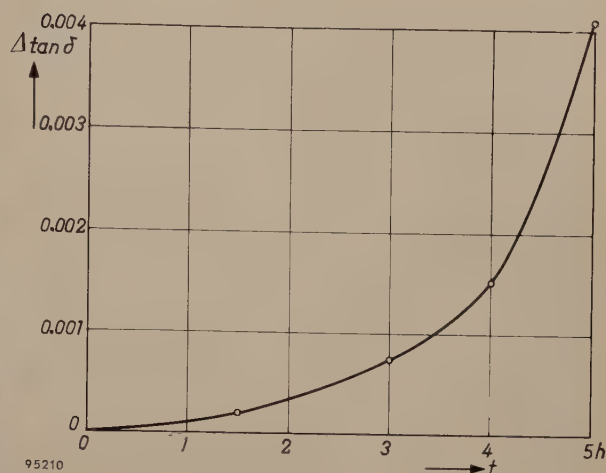


Fig. 7. Increase of loss factor  $\tan \delta$  measured after the dielectric had been loaded under  $42 \text{ V}/\mu$ , at room temperature, for a time  $t$  ( $3.8 \mu\text{F}$  capacitors having three layers of paper  $8 \mu$  thick, impregnated with pentachlorodiphenyl). To minimize the rise in dielectric temperature, the capacitors were loaded continuously for only half-hours at a time, with intermediate cooling.  $\tan \delta$  was measured at a dielectric temperature of  $120^\circ \text{C}$  to improve the sensitivity of the measurements.

by a variable alternating voltage in series with a choke  $L$ . The latter limits the current in the event of  $C_x$  breaking down during the measurement. By almost balancing the bridge the voltage across the other diagonal, in so far as the fundamental and the harmonics are concerned, is made very small. Apart from this small residual voltage there will be another, irregular, voltage present if ionization occurs in the capacitor. The total output voltage of the bridge is amplified, in which process components having frequencies below about  $8000 \text{ c/s}$  are removed. The result is made visible on an oscilloscope.

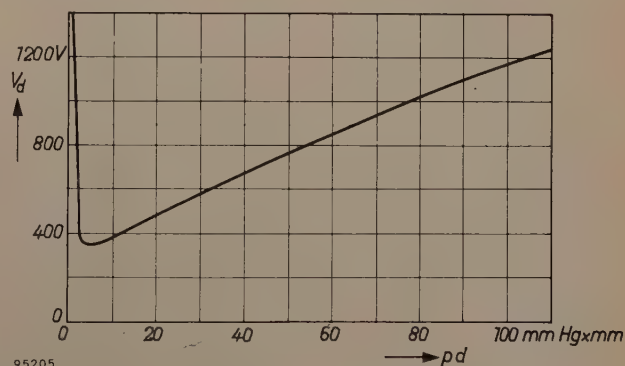


Fig. 8. The Paschen curve for air: breakdown voltage  $V_d$  for air in a uniform electrical field, as a function of the product  $pd$  of air pressure  $p$  (in mm Hg) and the distance  $d$  (in mm) between the electrodes.



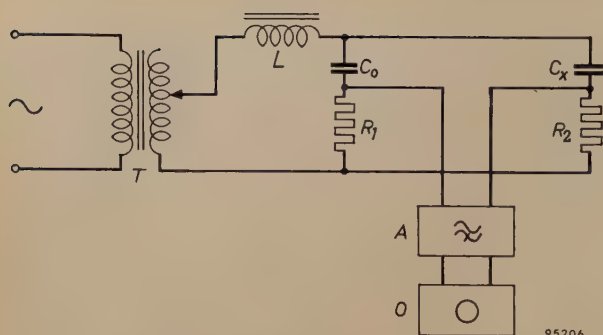


Fig. 9. Circuit for determining the degree of ionization in the dielectric of a capacitor. The test capacitor  $C_x$ , a high-quality and low-loaded capacitor  $C_0$  and resistors  $R_1$  and  $R_2$  form a bridge circuit. This is fed from the mains transformer  $T$  via choke  $L$ . The amplifier  $A$  transmits only components having frequencies above about 8000 c/s.  $O$  cathode-ray oscilloscope.

Some examples of the oscillograms so obtained are shown in fig. 10. The tests were made on non-impregnated capacitors. The dielectric consisted of three layers of paper 8  $\mu$  thick. After drying and degassing, the capacitors were filled with air at 1 atm instead of oil, and the cans were then sealed by soldering. After cooling to room temperature

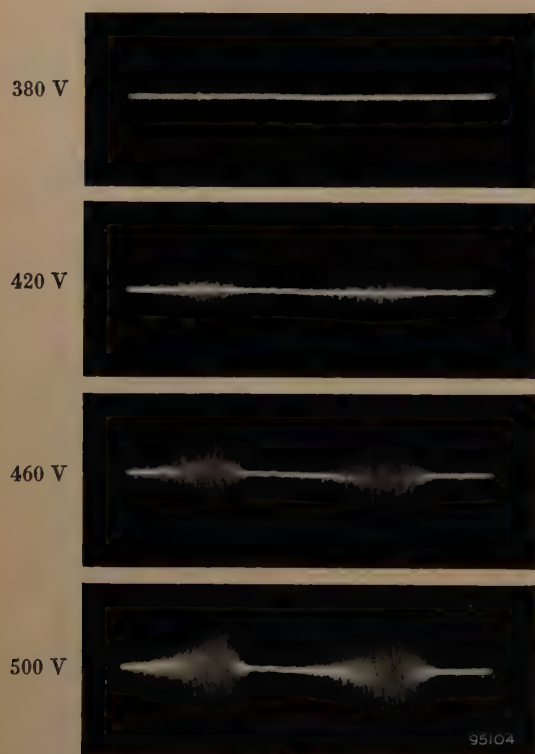


Fig. 10. Oscillograms obtained with the circuit of fig. 9. With 380 V across  $C_x$  the first small ionization peaks become visible. As the voltage is raised, the amplitude and number of the peaks increase rapidly.

The total length of the traces shown corresponds to 1/50 sec. The capacitor under investigation had a capacitance of about 2.5  $\mu$ F and a non-impregnated dielectric consisting of three layers of paper 8  $\mu$  thick.

the internal pressure was about 500 mm Hg. Since the non-impregnated dielectric contains innumerable gaps between the paper fibres, capacitors of this kind are eminently suitable for demonstrating ionization phenomena.

By placing such a capacitor under a bell jar, connected to a vacuum pump, and making a small opening in the can, the potential at which ionization begins can be found at different air pressures. The results are shown in fig. 11, curve 1. Like the curve representing Paschen's law (fig. 8), the curve found in this case shows a minimum; the numerical values, however, differ markedly from those with an arrangement of flat electrodes with only air between them.

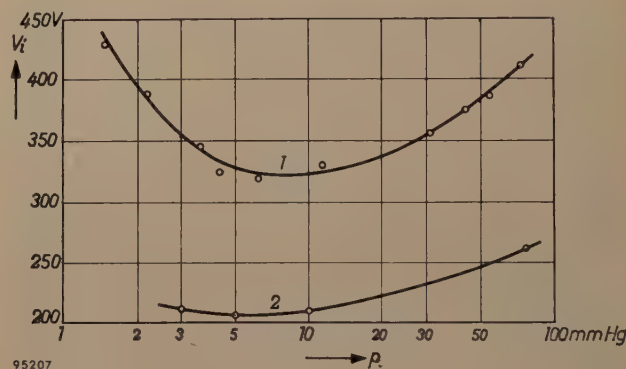


Fig. 11. Curve 1: voltage  $V_i$  at which ionization begins (in the capacitor referred to in fig. 10) as a function of the air pressure  $p$  in the capacitor. Curve 2 was measured after the capacitor had been impregnated with a mineral oil (the impregnation being deliberately poor).

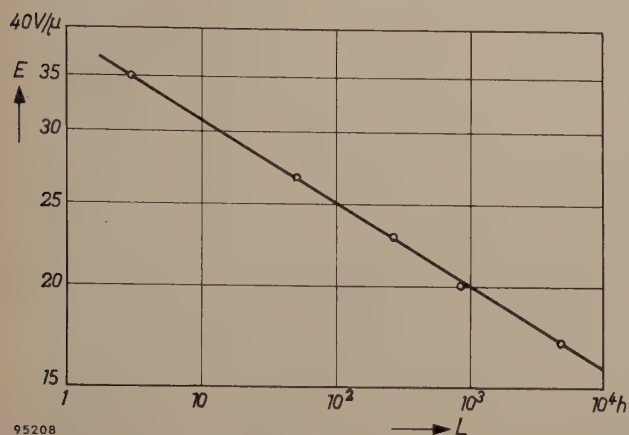
By properly impregnating the capacitor, one can make the ionization begin at a higher voltage than in a non-impregnated capacitor. A badly impregnated capacitor, on the other hand, can lead to ionization at a lower voltage, as low in fact as 200 V (fig. 11, curve 2).

In practice capacitors are impregnated at a high temperature, and the can is sealed at about 80 °C. During cooling to room temperature the pressure in the can decreases as a result of shrinkage of the impregnating fluid. The amount by which the pressure decreases depends on the temperature difference and on constructional details, such as the degree of filling and the stiffness of the can. This explains the unexpected fact (cf. fig. 5) that capacitors which have a reasonably long life at 80 °C and higher, frequently have a very short life at low temperature: the reduced pressure arising at low temperature is conducive to the formation of cavities in the dielectric, which are filled with gas at this low pressure. Considerably more ionization then occurs than at



higher temperatures, with adverse consequences for the life of the capacitor <sup>7)</sup>).

Some typical results of life tests are shown in *fig. 12*, which refer to capacitors of the same type as used for the tests of *fig. 5*. These capacitors, which



*Fig. 12.* Field strength  $E$  at which capacitors (of the same type as in *fig. 5*) reach a life  $L$  at an ambient temperature of about 25 °C. Each point represents the average of at least 20 individually-tested capacitors.

exhibit fairly heavy ionization, were tested at room temperature at different field strengths. The life is found to be inversely proportional to the tenth power of the field strength. Only at field strengths smaller than about 7 V/μ can the line be expected to level off towards longer life values, because with fields of this order it was no longer possible to detect any ionization.

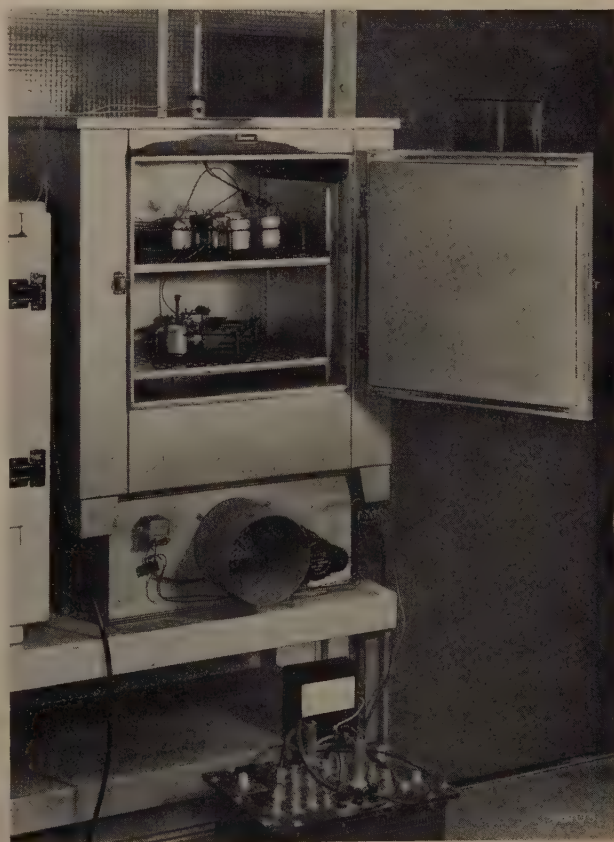
According to *fig. 5* it was to be expected that at 17.5 V/μ and room temperature the life of a capacitor would be far in excess of  $10^5$  hours (12 years). It can be seen from *fig. 12*, however, that, owing to ionization, the life is in fact only about 4400 hours.

#### Tests at periodically varying temperatures

Capacitors for the ballasts of gas-discharge lamps are normally intended to operate at voltages varying from 200 to 600 V. At these voltages the possibility of ionization must always be taken into account. A life test should therefore be carried out at the most unfavourable, i.e. the lowest, operating temperature that can occur. On the other hand, one must also reckon with the highest ambient temperature likely to be encountered. In this case, it is true, there will be less ionization or even none at all, but against this is the fact that the dielectric may deteriorate at high temperature and there is a possibility that thermal instability will set in.

<sup>7)</sup> In the case of high-power capacitors this can be avoided by mounting the capacitor elements in a steel cylinder. After evacuation and impregnation the cylinder is filled with a gas under high pressure (see article <sup>4)</sup>). This method is not suitable for low-power capacitors.

A satisfactory method of limiting the number of tests while still making full allowance for the various influences is to vary the temperature periodically. The procedure is as follows. The capacitors to be tested are placed in an oven and the capacitors and the oven are switched on simultaneously. Owing to the dielectric losses and the rising temperature of the oven the capacitors get hot, in a manner corresponding to practical conditions. Some time after temperature equilibrium has been reached, for example after 4 hours, the oven as well as the capacitors are switched off. Since the oven cools much more slowly than a capacitor under practical conditions, a fan is switched on at the same moment in order to accelerate the cooling. The oven and the capacitors are subsequently switched on again, and the process is repeated. A photograph of the apparatus is shown in *fig. 13*. The equipment is suitable for examining capacitors that are intended for indoor lighting installations, and which are therefore not exposed to particularly low temperatures. The upper limit of the alternating oven-temperature can be fixed to correspond to the highest ambient temperature to



*Fig. 13.* Apparatus for performing life tests with periodically varying ambient temperature. The capacitors are arranged on racks to ensure that their mutual spacing remains the same in every test. A fan (under the oven) blows air at about 20 °C through the oven during the cooling process.



which the capacitors are likely to be exposed in actual operation (account being taken of the heating effect of lamps and chokes or transformers in the lamp fitting).

Capacitors intended for outdoor installations, for example for road lighting, must also be able to withstand temperatures as low as  $-20^{\circ}\text{C}$ . In this case the lower limit of the temperature range is obtained with a refrigerating system.

To shorten the duration of the test the field strength can be boosted to a value higher than the maximum permissible value under operating conditions. The test can be carried out at several values of field strength, and the life at the normal field strength predicted by extrapolating the results. An example is given in *fig. 14*. This relates to capacitors of the

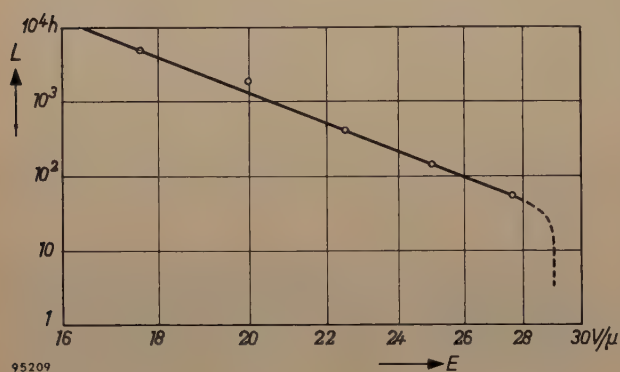


Fig. 14. Life  $L$  of capacitors (the same type as in *figs. 5* and *12*) in an alternating-temperature test as a function of field strength  $E$ . The temperature limits were  $20$  and  $80^{\circ}\text{C}$ . Each point represents the average for 18 capacitors.

At  $E \approx 29 \text{ V}/\mu$  the capacitors enter into the unstable region during the warm part of the cycle. At this point the life curve may therefore be expected to bend over to very short life values.

same type as those of *fig. 12*. They were loaded under electric fields of various values, at temperatures varying from  $20$  to  $80^{\circ}\text{C}$ . The points found lie almost on a straight line which runs approximately parallel to the line shown in *fig. 12*, but is displaced somewhat towards longer life values. This is quite understandable, for condensers of the type in question are most heavily loaded at low temperature. In the test this happens for only a part of the time; the higher temperature prevailing during the remainder of the test caused little damage in this case, so that a longer life was found than would have been the case at a constantly low temperature.

### High-voltage breakdown tests

The high-voltage test is a familiar and quick method of examining capacitors, and one which

is found in many inspection specifications. The test is sometimes carried out with direct voltage, sometimes with alternating voltage.

In the first case only the breakdown potential is determined, or at least whether this voltage is higher than a specific value. It provides no information whatsoever on the much more important properties of the dielectric, such as the power factor, the likelihood of ionization and the dependence of the aging of the dielectric on temperature.

The test with alternating voltage seems to be more useful in that dielectric losses and ionization are not entirely neglected. During the short duration of the test, however (usually one minute), the capacitor does not become hot enough to allow thermal instability, which is almost certainly present, to be detected. Nor does the ionization have any opportunity to cause perceptible damage.

Neither of these tests, therefore, discloses more than the capacitor's ability to withstand peak voltages of short duration (and the same applies to the surge-voltage tests specified in some countries). Nothing can be concluded from these about the life of the capacitor under practical conditions. It is a simple matter, for example, to make a capacitor capable of withstanding a high test voltage but which, in actual operation, will not have a long life. All that is necessary for this purpose is to build up the dielectric from, say, four layers of paper  $7.5 \mu$  thick, and to choose a type of paper and impregnating agent having high losses which rise steeply with temperature. Because of the division into four layers there is very little chance that weak spots in the paper will overlap. It is therefore readily possible to attain a breakdown voltage of  $6000 \text{ V}$ , which is 15 times the value of a reasonable operating voltage for a dielectric with a total thickness of  $30 \mu$ . At quite a low ambient temperature, however, the capacitor will no longer be thermally stable. If one modifies the design by taking two layers of paper  $15 \mu$  thick, the breakdown voltage will drop to about  $3000 \text{ V}$ , but if the paper and impregnating agent are of good quality one will now have a capacitor with much greater thermal stability and a longer life.

In order, therefore, to properly assess the life of capacitors it is necessary to subject them to life tests under conditions of reasonable field strength overload — e.g. not exceeding 1.5 to 2 times the normal field strength — and to vary the temperature periodically between limits equal to those which will be encountered in practice.

If capacitors for lamp ballasts are developed and tested in this way, their life will not be inferior to that of transformers and chokes. The view still



widely held that the capacitor is the weakest link in the chain may once have been justified, but nowadays it need by no means be so.

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**Summary.** In ballasts for gas-discharge lamps capacitors are often used with the object, among other things, of improving the power factor. The life of these condensers depends not only on temperature (as in the case of transformers and chokes) but also on the field strength.

Associated with the fact that the dielectric losses increase sharply with temperature is the danger of thermal instability: if the ambient temperature exceeds a certain critical value (the stability limit) a stable temperature state is no longer attained and the capacitor soon breaks down.

The higher the ambient temperature the quicker the dielectric's reserve of insulation is exhausted — analogous to what is found with the insulation of transformers. This suggests that the capacitor should have an extremely long useful life at low ambient temperature. However, the lower pressure obtaining in sealed capacitor cans increases the chance of gas bubbles forming in the dielectric and of ionization occurring in these bubbles. This ionization causes the dielectric to age very rapidly. The author discusses a method of demonstrating the presence of ionization.

The familiar high-voltage breakdown test merely discloses whether the breakdown potential exceeds a certain value, and provides no information on the expected life of the capacitor. An efficient method of life testing is to load the capacitors with a field strength 1.5 to 2 times higher than normal, while at the same time periodically varying the ambient temperature between the extreme values to which the capacitors might be exposed in actual operation.

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## ANNEALING TELEVISION PICTURE-TUBE BULBS



The bulb of a television picture-tube is made up of three parts: the conical body, the cylindrical neck and the window. When these parts are fused together, stresses are set up in the glass. To relieve these stresses the bulbs are placed on a conveyer belt which passes them through a tunnel annealing oven. The above photograph, taken in the new television tube factory in Eindhoven, shows this process in operation.



## A SIMPLE AND COMPACT ARRANGEMENT FOR MEASURING THE $\beta$ -ACTIVITY OF WEAK RADIOACTIVE SAMPLES

621.387.4:539.16.08

In recent years greatly increased importance has been attached to the measurement of very weak radiations, particularly in connection with the radioactive contamination of the atmosphere, rainwater, foodstuffs and the waste water from medical, radiochemical and nuclear-physics laboratories. For all such purposes, detectors are required that combine extreme sensitivity with the utmost reliability.

In the majority of cases the aim is to monitor the beta activity, for example of the hazardous isotope  $\text{Sr}^{90}$ . The Geiger counter tube with end-window, such as the Philips types 18505 and 18506, is still the most commonly used beta detector, because of its high sensitivity to beta rays, its great stability, and the simplicity of the auxiliary equipment<sup>1)</sup>.

Among the most important factors governing the lower limit of detection are the *background counts* of the detector. The background count is the number of discharges in the tube per unit time caused by any agency other than that which is to be detected. They are caused, *inter alia*, by:

- 1) gamma radiation originating from the surroundings, e.g. from neighbouring radioactive preparations, natural radioactivity of building materials (in which the isotope  $\text{K}^{40}$ , for instance, is often present), or from the air (which may carry radon or disintegration products of radon);
- 2) mesons of cosmic radiation;
- 3) beta and gamma radiation originating from impurities in the materials of which the counter tube is made.

The interference from neighbouring gamma radiation can be eliminated by shielding. Obviously, the shielding materials must themselves contain no radioactive impurities, particularly that part of the shielding in the immediate vicinity of the counter tube. Lead has generally too much radioactive contamination for the purpose. Iron is more satisfactory, but best of all is mercury, because it can readily be purified. Reasonable shielding is possible with a mercury shield of, say, 3 cm thickness, enclosed in a 15 cm thick iron shield; the mercury then absorbs the radiation from the radioactive impurities in the iron.

Owing to the very great penetrating power of mesons, other means must be sought of overcoming

this source of error. Hitherto it has been customary to surround the counter tube with a number of *guard counters*, which are so disposed that every meson triggering the central counter necessarily triggers one or more guard counters also. The circuit operates on the anti-coincidence principle, that is to say a count from the central tube is recorded only when none of the guard counters are simultaneously triggered.

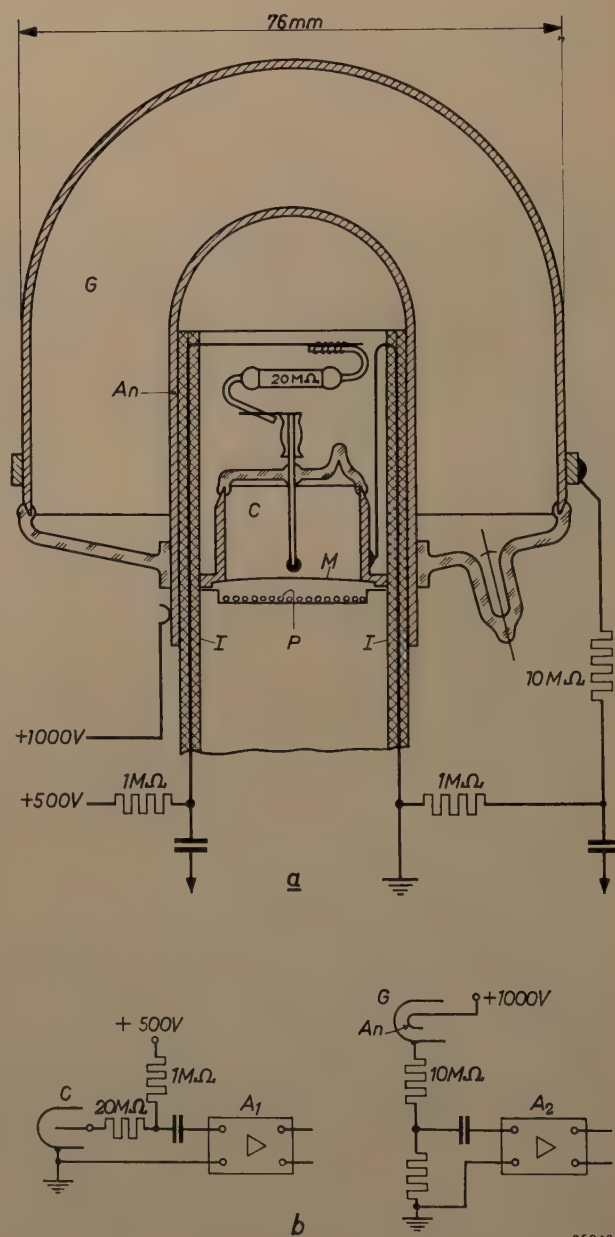


Fig. 1. a) Cross-section and b) circuit of a guard counter  $G$  with hollow anode  $A_n$ , in which is situated the central beta counter  $C$  (with mica window  $M$ ).  $I$  insulation.  $P$  radioactive sample.  $A_1$  and  $A_2$  amplifiers.

<sup>1)</sup> See N. Warmoltz, Geiger-Müller counters, Philips tech. Rev. 13, 282-292, 1951/52.



Such an arrangement, consisting of 10 to 15 counter tubes, is naturally fairly voluminous. If it is surrounded by a 3 cm shield of mercury plus a 15 cm shield of iron to screen it from gamma radiation, the aggregate equipment weighs something like

The fact of there only being one guard counter offers the following advantages:

- 1) the smaller size entails a great saving in the weight of the shielding (a saving of more than 1000 kg),

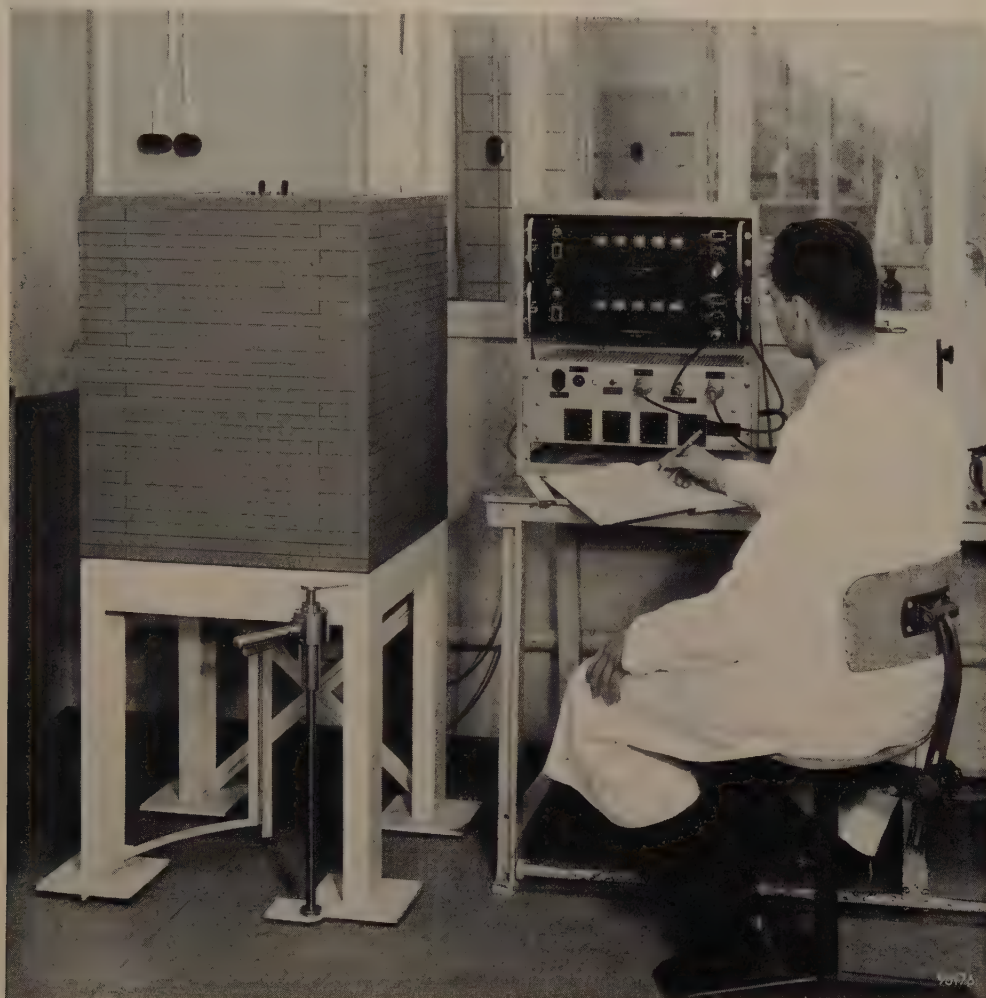


Fig. 2. Complete equipment for measuring weak beta activity. Left, gamma-ray shielding, consisting of 20 cm thick iron blocks surrounding a 2.5 cm thick shield of mercury. The latter surrounds an arrangement of a counter tube and guard counter as illustrated in fig. 1. Right, the amplifiers in an anti-coincidence arrangement, on top of which are mounted two scalars, each composed of five decimal counter tubes type EIT<sup>2</sup>). The number of counts before and after meson correction can be read directly from the tubes in the front panel.

<sup>2</sup>) Philips tech. Rev. 14, 313, 1952/53.

2000 kg, which is too cumbersome in many cases.

Fig. 1 illustrates an arrangement of counter tubes using a new type of guard counter. The two electrodes of the guard counter are concentric hemispheres to which coaxial cylinders are connected. The beta counter fits inside the hollow anode of the guard counter. With this arrangement no meson can actuate the inner counter tube without at the same time actuating the exterior counter. In its operation this tube is similar in many respects to the counter tubes described in an earlier publication by the Philips laboratory, Amsterdam<sup>3</sup>). The complete monitoring equipment is shown in fig. 2<sup>4</sup>).

- 2) the layout and supervision of the equipment are greatly simplified, and
- 3) the electronic apparatus is simpler.

The central counter tube in fig. 1 is a modified experimental version of type 18505. The main differences are:

- <sup>3</sup>) J. Hermesen, A. M. J. Jaspers, P. Kraayeveld and K. van Duuren, New Geiger tube designs, hollow anode and parallel plate counters, Proc. International conference on the peaceful uses of atomic energy, Geneva 1955 (published by United Nations, New York 1956), part 14, pages 275-276.
- <sup>4</sup>) A more detailed description of this measuring arrangement was given by K. van Duuren, W. K. Hofker and J. Hermesen at the 1958 Geneva Conference (to be published as contribution P. 2343 in the Proceedings of this conference).



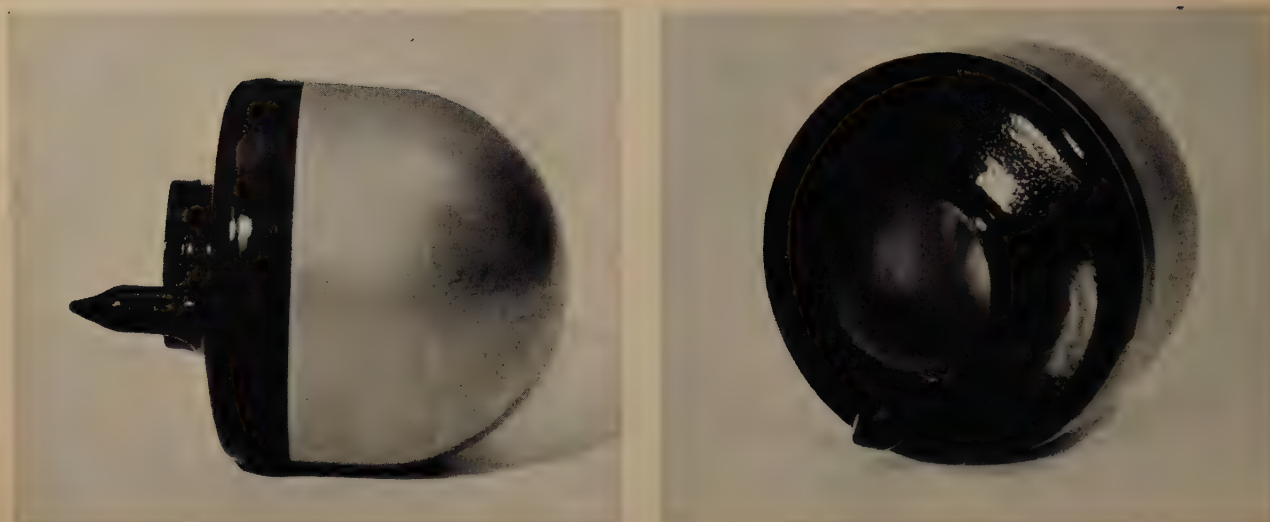


Fig. 3. Guard counter as in fig. 1.

- a) the length has been cut down to  $1/3$  of the original length, to reduce the tube's sensitivity to gamma radiation;
- b) the materials used have been carefully chosen to keep the contribution made by the tube wall to the background counts as low as possible.

In a particular case the background counts of the counter tube inside the guard counter were reduced

to 0.7 per minute, as compared with the 20 background counts recorded in the same laboratory with a normal counter tube, type 18505, having the same sensitivity to beta rays but without shielding.

Photographs of the guard counter tube are shown in fig. 3.

K. van DUUREN \*).

\*) Philips laboratory (Synchrocyclotron), Amsterdam.

## ABSTRACTS OF RECENT SCIENTIFIC PUBLICATIONS BY THE STAFF OF N.V. PHILIPS' GLOEILAMPENFABRIEKEN

Reprints of these papers not marked with an asterisk \* can be obtained free of charge upon application to Philips Electrical Ltd., Century House, Shaftesbury Avenue, London W.C. 2.

**2568\***: H. Bremmer: Comparaison des fluctuations de l'amplitude et de la phase des champs engendrés par diffusion (champs de turbulence) (*Onde élect.* **37**, 498-500, May 1957; in French).

This article deals with the statistical theory of fading phenomena, in particular with respect to the propagation by scattering of microwaves. The fading occurring at large distances (far beyond the horizon) is compared with that within the horizon. In both these cases special attention is paid to the difference in fading rate for the amplitude and that for the phase. These two fading rates differ at large distances; for short-distance scattering they are equal as far as the approximations used here are concerned.

**2569\***: N. Warmoltz and H. A. M. de Grefte: Spectromètre de masse pour détection des fuites, fonctionnant avec un mélange à

faible teneur en hélium (*Le Vide* **12**, 202-207, 1957, No. 69; in French).

A mass spectrometer leak detector of the Nier type with a radius of curvature of 5 cm is described using an electron multiplier for measuring the ion current. In this manner the sensitivity is increased so far that one can use as probe gas for leak testing a mixture containing only 1% of helium. The apparatus can also be used as an analyser for masses up to about 50.

**2570**: J. P. M. Gieles: The measurement of group delay in triode amplifiers at 4000 Mc/s (*Onde élect.* **37**, 781-788, October 1957).

In microwave repeaters for long distance links using frequency modulation, not only the amplitude characteristic but also the phase characteristic is extremely important. The latter can be described in terms of the group delay, which should be as constant as possible over the transmission band



desired. The total delay variation admissible for colour television could perhaps be as low as 50 m $\mu$ s. For direct UHF links equipped with EC 56, this results in a variation of less than 0.1 m $\mu$ s per stage if the distortion is cumulative, so that a very high accuracy in the measurement of the group delay is necessary. Measurements are performed in a closed loop that is made to oscillate by the amplifier under test. The loop also contains a precision phase shifter. A simple formula can be derived, expressing the total group delay of the loop in terms of the frequency shift resulting from an introduced phase shift. The frequency shift can be measured very accurately by a beat-tone procedure. The results have an accuracy of about 0.1 m $\mu$ s and indicate that the triode is indeed capable of fulfilling the severe requirements. Finally, some remarks are made concerning level to phase conversion.

**2571:** J. Links, J. E. Rombouts and P. Keulen: The "bulging factor", a fungistatic antibiotic produced by a streptomyces strain, with evidence of an active water-excreting mechanism in fungi (J. gen. Microbiol. **17**, 596-601, 1957, No. 3).

The "bulging factor", a new antibiotic with a chemical nature related to streptothricin, is produced by a Streptomyces strain and was isolated as reineckate and picrate. A study on its mode of action resulted in the development of a plate technique for assaying bulging factor activity. Evidence is presented in support of a theory about the existence of an active water-excreting mechanism in fungi susceptible to the bulging factor.

**2572\*:** G. Ahsmann: On the impedance of the anode-fall region of a glow discharge in the noble gases (Rendiconti Terzo Congresso internazionale sui fenomeni d'ionizzazione nei gas, Venice 11-15 June 1957, Fondazione "Giorgio Cini", Società Italiana di Fisica, Milan 1957, pp. 19-25).

The complex impedance of glow discharges was measured in a discharge tube fitted with adjustable electrodes. As the electrode distance is increased the impedance is initially independent of the distance; however when the distance is so great that an anode fall begins to develop, the impedance in a given frequency region increases rapidly. The cause of this increase lies in the anode fall layer and in the Faraday dark space. An equivalent circuit is given for the anode region of the discharge when an anode fall is present.

**2572a\*:** J. A. Kok: Mechanism and properties of various types of electrical discharges in gas-filled triodes (Rendiconti Terzo Congresso internazionale sui fenomeni d'ionizzazione nei gas, Venice 11-15 June 1957, Fondazione "Giorgio Cini", Società Italiana di Fisica, Milan 1957, pp. 548-557).

A report on the work described here has also appeared in Appl. sci. Res. **B5**, 445-553, 1956 and **B6**, 207-221, 1956; see these abstracts Nos. **2368** and **2445**.

**2572b\*:** W. Verwey: Probe measurement of electron temperature in the positive column of a rare-gas discharge and correlation with microwave noise (Rendiconti Terzo Congresso internazionale sui fenomeni d'ionizzazione nei gas, Venice 11-15 June 1957, Fondazione "Giorgio Cini", Società Italiana di Fisica, Milan 1957, pp. 1115-1130).

The Langmuir probe is a very useful tool for the determination of electron temperatures in the positive column of gas discharges. Its reliability however, is subject to some doubt, especially at high current densities and comparatively high gas pressures. This article describes measurements with the probe on rare-gas discharge tubes at pressure up to 20 mm Hg, and for current densities of 0.04 A/cm<sup>2</sup>. These tubes were developed as standard noise sources for the cm waveband. The electron temperatures measured with the probe are found to be in good agreement with those determined from noise measurements. In the probe measurements special attention was paid to the effect of the work function of the probe material.

**2572c\*:** N. Warmoltz and H. J. J. van Boort: A flash tube for colour photography of the human retina (Rendiconti Terzo Congresso internazionale sui fenomeni d'ionizzazione nei gas, Venice 11-15 June 1957, Fondazione "Giorgio Cini", Società Italiana di Fisica, Milan 1957, pp. 1131-1134).

Short description of a method of making colour photographs of the human retina for diagnostic purposes. A special electronic flash tube has been developed that has a very high brightness (peak value  $\sim 1.5 \times 10^6$  cd/cm<sup>2</sup> ( $9 \times$  brighter than the sun) and a flash duration of 3.5 msec. With this very short exposure (which is sufficient because of the high brightness) sharper photographs are possible than with the usual carbon arc. A 6000  $\mu$ F condenser charged to 500 V dissipates an energy of 750 joules in the lamp. See also Philips tech. Rev. **15**, 342-346, 1953/54.



**2573:** J. Smit: The spontaneous Hall effect in ferromagnetics, II (Physica **24**, 39-51, 1958, No. 1).

It is shown that the spontaneous part of the Hall effect arising from the spontaneous magnetization is caused by skew scattering of the magnetized conduction electrons (in this case the 3d-electrons) due to their transverse polarization induced by spin-orbit interaction, which acts as an impact parameter in the collision process. The problem could be approximately solved for scattering on an impurity atom by converting it into that of the scattering on a vortex-like perturbing centre. The order of magnitude of the calculated Hall angle agrees with the experimental one ( $\approx 10^{-2}$ ). It is shown quite generally that for lattice vibration scattering the Hall effect should be proportional to the square of the resistivity, as has been found experimentally for pure iron and nickel.

**2574:** U. Enz: Time-dependent constricted hysteresis loops in a single crystal of manganese ferrous ferrite (Physica **24**, 68-70, 1958, No. 1).

The time-dependence of the initial permeability (i.e. disaccommodation) is measured on a single-crystal toroid of manganese ferrous ferrite. This effect is as high as 95% of the value immediately after demagnetization of the ring. It is shown that disaccommodation is a time-dependent blocking of domain walls. The blocked walls cause constricted minor hysteresis loops, characterized by a critical magnetic field which is necessary to overcome the blocking force. The constriction is time-dependent.

**A 2:** E. Kauer and A. Rabenau: Über den Bandabstand von Galliumnitrid und Aluminiumnitrid (Z. Naturf. **12a**, 942-943, 1957, No. 11). (On the band structure of gallium nitride and aluminium nitride; in German.)

Note on the determination of the energy difference between the conduction band and the valency band in polycrystalline gallium nitride and aluminium nitride, by means of measurements of the diffuse reflection as a function of the wavelength of the incident radiation. The (room-temperature) values found are: for gallium nitride 3.2 eV and for aluminium nitride  $>5$  eV.

**A 3:** A. Rabenau and P. Eckerlin: Die  $K_2NiF_4$ -Struktur beim  $La_2NiO_4$  (Acta cryst. **11**, 304-306, 1958, No. 4). (The  $K_2NiF_4$  structure in  $La_2NiO_4$ ; in German.)

Report of an X-ray diffraction investigation into the structure of  $La_2NiO_4$  using Debye-Scherrer photographs and Cu  $K\alpha$  radiation. It is found that

$La_2NiO_4$  has a structure related to the perovskite structure already found in  $K_2NiF_4$  and studied by Balz and Plieth. The lattice constants of the tetragonal  $La_2NiO_4$  were found to be:  $a = 3.855 \pm 0.001$  Å and  $c = 12.652 \pm 0.003$  Å. The  $K_2NiF_4$  structure is also exhibited in  $La_2CoO_4$ , but here it was orthorhombically deformed. The lattice constants are:  $a = 5.539$  Å,  $b = 12.66$  Å,  $c = 5.482$  Å.

**A 4:** K. Jost and G. Schiefer: Die Auswertung von Vierpol- und Materialmessungen mit dem logarithmischen Leitungsdiagramm (Archiv elektr. Übertragung **12**, 295-300, 1958, No. 7). (The analysis and evaluation of 4-pole and material measurements with the aid of the logarithmic transmission-line chart; in German.)

A method is described that allows the quadrupole parameters, and in particular the material constants  $\mu$ ,  $\epsilon$ ,  $\tan \delta_\mu$  and  $\tan \delta_\epsilon$  to be measured with the aid of a coaxial slotted line. The numerical evaluation is greatly simplified by the adoption of a logarithmic transmission-line chart whose curves of constant VSWR and constant node displacement  $l/\lambda$  simultaneously represent curves of the function  $\ln \tanh(a + jb)$  for constant  $a$  and  $b$ .

**R 339:** S. Duinker: General properties of frequency-converting networks (Philips Res. Repts. **13**, 101-148, 1958, No. 2).

Continuation of **R 337**.

**R 340:** H. L. Spier and W. L. Wanmaker: Influence of additives on particle size of tungsten powder prepared by reduction with hydrogen from tungsten trioxide (Philips Res. Repts. **13**, 149-156, 1958, No. 2).

An investigation is carried out into the reduction with hydrogen of tungstic acid and tungsten trioxide doped with various additives. It is shown that the addition of potassium silicate and potassium silicotungstate results in a marked growth of the tungsten particles, while other additives as calcium nitrate or magnesium sulphate tend to give a small particle size. From the behaviour of the pure potassium silicotungstates during reduction, an explanation of the observed phenomena is proposed.

**R 341:** W. van Gool: Fluorescence and photoconduction of silver-activated cadmium sulphide (Philips Res. Repts. **13**, 157-166, 1958, No. 2).

Several Ag-activated CdS phosphors are described, with Ga or Cl as coactivator. The fluorescence at low temperature shows two bands with maxima at



6200 Å and 7300 Å. High Ag concentration and low coactivator concentration promote the short-wave emission. Phosphors with equal concentrations of activator and coactivator show only the long-wave emission. Optimal photoconduction and low dark-current are only found with an activator-coactivator ratio slightly greater than unity. A series of mixed crystals (Zn,Cd)S activated with Ag shows clearly the relation between 6200 Å emission in CdS and a 3880 Å emission in ZnS and also between the 7300 Å band in CdS and the normal blue Ag band at 4350 Å in ZnS. The behaviour of the Ag bands in CdS is quite similar to those of Cu in ZnS. These results show that the conclusions drawn by Lambe and Klick regarding the position of the impurity level responsible for the 6200 Å Ag emission in CdS cannot be applied to the normal blue Ag emission in ZnS-Ag. These results led to an improvement of the properties of a red colour-television phosphor.

**R 342:** J. Bloem: Discussion of some optical and electrical properties of  $\text{Cu}_2\text{O}$  (Philips Res. Repts. **13**, 167-193, 1958, No. 2).

From known optical and electrical data on  $\text{Cu}_2\text{O}$  a band scheme is constructed for this semiconductor. The equilibria between the solid constituents and a vapour containing oxygen are calculated. From these data the conductivity and the concentrations of the various defect centres can be evaluated for  $\text{Cu}_2\text{O}$  at high temperatures in equilibrium with an applied partial pressure of oxygen. The results are shown to be in good agreement with experiment. The optical and electrical properties after quenching, however, do not correspond to the model mentioned above. If association between defects is considered during the cooling process, the optical properties can be explained. As to the Hall-effect data, the occurrence of chemisorption of oxygen during quenching must also be taken into account, especially if well-conducting surface layers are formed, which appears to be the case at the lower oxygen pressures preferently, whereas at high oxygen pressures conduction through the bulk of the crystallites prevails. The intermediate region of mixed conduction is characterized by a very low effective mobility.

**R 343:** K. M. Adams: On the synthesis of three-terminal networks composed of two kinds of elements (Philips Res. Repts. **13**, 201-264, 1958, No. 3).

In this thesis (Delft, 1957) the synthesis of series-parallel LC three-terminal networks is investigated.

A set of necessary and sufficient conditions and a method of realization of all sets of series-parallel LC three-terminal network functions from the zeroth to the sixth degree are given. Some of these conditions are essentially new, that is, independent of any previously derived conditions. They are necessary for the synthesis of series-parallel LC three-terminal networks, but it is not known whether they are also necessary for the synthesis of three-terminal networks of arbitrary structure. In principle, it is possible to apply the method to functions of higher degree than the sixth, but the amount of computation required increases in general very rapidly with the degree.

**R 344:** W. Ch. van Geel, C. A. Pistorius and P. Winkel: Photo effects with anodic oxide layers on tantalum and aluminium (Philips Res. Repts. **13**, 265-276, 1958, No. 3).

This article deals with the photoelectric properties of the system  $\text{Ta-Ta}_2\text{O}_5$ -electrolyte during irradiation with ultraviolet light. The electrolyte has also been replaced by a transparent layer of a metal or a semi-conductor. When the system is short-circuited, a photo-current ( $i_0$ ) is produced which is proportional to the intensity of irradiation. With a high external resistance a photo-e.m.f.  $V$  appears which shows considerable inertia as a function of the time  $t$  and which can be written as  $V = V_{\max}(1 - e^{-kt})$ . It appears that  $k$  is proportional to the intensity of irradiation. The value of  $k$  decreases with increasing thickness  $d$  of the layer. After correcting for this change of  $k$ , which is probably due to absorption, we find that  $i_0 d = \text{constant}$ . An attempt is made to explain the observed phenomena by assuming that the work function  $\text{Ta-Ta}_2\text{O}_5$  is smaller than the work function electrolyte- $\text{Ta}_2\text{O}_5$ .

**R 345:** P. Winkel, C. A. Pistorius and W. Ch. van Geel: On the relation between current and field during anodic oxidation (Philips Res. Repts. **13**, 277-295, 1958, No. 3).

Starting from the assumption that an anodic oxide layer on metals like Al and Ta has an amorphous structure, a relation between field and current during anodic oxidation is derived. It appears that this relation is somewhat different from the exponential one usually derived in literature (Verwey, Mott). With the new relation, it is possible to explain the various results obtained with the experiments presented in this paper. Probably it may explain the rather different numerical results mentioned in literature in the same way. The consequences of the amorphous structure on a more detailed picture



of the oxidation process have also been calculated. The results point to a frequency dependence of one of the parameters as was actually observed in the experiments. The observed agreement between theory and experiments has made it possible to calculate approximately the parameters introduced in this theory.

- R 346:** C. Z. van Doorn: Quantum efficiency of *F*-centre fluorescence in KCl (Philips Res. Repts. 13, 296-300, 1958, No. 3).

The quantum efficiency of *F*-centre fluorescence at 77 °K in X-ray and additively coloured KCl has been measured using an integrating sphere. Values of up to 91% have been found, depending on the concentration of *F* and *M* centres.

- R 347:** A. van Weel: Analysis of diode-detector circuits for signals with asymmetrical sidebands (Philips Res. Repts. 13, 301-326, 1958, No. 4).

A theoretical analysis is given of a diode detector stage for a modulated signal with asymmetrical sidebands of which the carrier is detuned with respect to the resonance frequency of the I.F. circuit. The equivalent circuit is found to be a three-port consisting of the impedances for upper- and lower-sideband frequencies and video frequency in series. Measurements show different overall characteristics in the case of equal positive or negative detuning of the carrier frequency; this effect is caused by the asymmetrical shape of the diode-current peak.

- R 348:** K. F. Niessen: Non-magnetic ions in an antiferromagnetic (Philips Res. Repts. 13, 327-334, 1958, No. 4).

An experimental method is described for determining the distribution of a relatively small number of foreign non-magnetic ions between the two sub-lattices of an antiferromagnetic, in which they replace the same number of original magnetic ions. The influence of the foreign ions on several measurable quantities is investigated. This influence can be used to determine some specific constants of the antiferromagnetic. In the anisotropy energy besides the usual terms an interaction term between the two sub-lattices is taken into account. Use is made of the spontaneous magnetization, the parallel and perpendicular susceptibilities, the critical field strength and the antiferromagnetic resonance.

- R 349:** H. J. Oskam: Microwave investigation of disintegrating gaseous discharge plasmas

(Philips Res. Repts. 13, 335-400, 1958, No. 4).

The disappearance of electrons from an isothermal disintegrating gaseous discharge plasma is investigated both theoretically and experimentally by considering the shift of the resonance frequency of a microwave cavity enclosing the plasma. The first few sections discuss theoretically such items as (1) the complex conductivity of a plasma at high frequencies, (2) the influence of the various loss processes on the shape of the electron density – time curve, (3) the connection between the conductivity of the plasma and the properties of the cavity for various types of cavity. In measuring the frequency shift as a function of time, it proves to be necessary to limit the power of the probing signal (frequency  $\approx 10000$  Mc/s) to a few microwatts. The construction and preparation of the gas containers and the measurements are presented. The disappearance process of the electrons from the plasma in helium is found to be ambipolar diffusion even at a pressure of 25 mm Hg. The mobility of the  $\text{He}_2^+$  ions in helium at standard density is  $\mu_0(\text{He}_2^+) = 17.3 \pm 0.7$  cm/s per volt/cm. In the neon afterglow, however, the electrons disappear at a pressure of 20 mm Hg by dissociative recombination with  $\text{Ne}_2^+$  ions ( $\alpha \approx 2.5 \times 10^{-7}$  cm<sup>3</sup>/s). An admixture of rare-gas atoms with a lower ionization potential than that of the main gas is found to have a great effect on the disappearance of the electrons. The afterglow is studied in the binary gas mixtures of helium with a small concentration of neon, argon, or krypton, and also in neon-argon and neon-krypton mixtures. Measurements in these mixtures all show the production of a considerable number of atomic ions of the admixture, even at very low concentrations of the latter. The process concerned in helium-neon is a charge-transfer process between a  $\text{He}_2^+$  ion and a neon atom; the relevant cross-section is found to be  $Q_{ce} \approx 1.5 \times 10^{-15}$  cm<sup>2</sup>. In the other mixtures the atomic ions are produced by the Penning effect and possibly by the above type of charge-transfer process, which cannot be separated in the present experiment. In all gas mixtures the atomic ions are found to be converted into molecular ions by three-body collisions with an atom of the main gas and an atom of the admixture. The probability of this conversion process proves to depend strongly on the difference between the ionization potential of the main gas and that of the admixed atoms, and is found highest in helium-neon. Moreover, the measurements suggest the conversion of  $\text{Ne}^+$  ions into  $(\text{HeNe})^+$  ions by a three-body collision with two helium atoms.